General Nonlinear 2-Fluid Hydrodynamics of Complex Fluids and Soft Matter

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Abstract

We discuss general 2-fluid hydrodynamic equations for complex fluids, where one kind is a simple Newtonian fluid, while the other is either liquid-crystalline or polymeric/elastomeric, thus being applicable to lyotropic liquid crystals, polymer solutions, and swollen elastomers. The procedure can easily be generalized to other complex fluid solutions. Special emphasis is laid on such nonlinearities that originate from the 2-fluid description, like the transport part of the total time derivatives. It is shown that the proper velocities, with which the hydrodynamic quantities are convected, cannot be chosen at will, since there are subtle relations among them. Within allowed combinations the convective velocities are generally material dependent. The so-called stress division problem, i.e. how the nematic or elastic stresses are distributed between the two fluids, is shown to depend partially on the choice of the convected velocities, but is otherwise also material dependent. A set of reasonably simplified equations is given as well as a linearized version of an effective concentration dynamics that may be used for comparison with experiments.

1 Introduction

The thermodynamic and hydrodynamic properties of multi-component complex fluids are determined by the microscopic degrees of freedom of their constituents and the coupling between these degrees of freedom. Such systems can exhibit rather rich phase behavior and dynamics, especially when one or more components is a structured or macromolecular fluid [1]. Due in part to the coupling of internal degrees of freedom, these systems can also exhibit novel flow-induced structural evolution phenomena, including shear-induced phase transformations and flow alignment of constituents on microscopic to mesoscopic length scales. Such structural evolution in turn leads to nonlinear rheological behavior, such as stress overshoots in response to imposed rates of strain, plasticity, and thixotropy.

The overwhelming complexity of the microscopic description of these systems, such a detailed description is often not well suited for analysis of the macroscopic dynamical behavior. Instead, explicit macroscopic models have been developed for this purpose. Some such models have been obtained by a suitable coarse-graining procedure starting from a microscopic theory. Others are purely phenomenological models constrained only by conservation laws, symmetry considerations and thermodynamics. The so-called "two-fluid" models for binary systems of distinct components or phases are useful examples of such a macroscopic approach [2]. In the two-fluid description, each component or phase is treated as a continuum described by local thermodynamic variables (e.g. temperature, density, and relevant order parameters), and dynamical quantities (e.g. velocity or momentum). In general, these variables for the constituents are coupled. For instance, the effective friction between components in a binary fluid mixture leads to a drag force in the macroscopic description that is proportional to the local velocity difference.

Two-fluid models have been employed in many different physical contexts. The two-fluid approach is a key element of many traditional models for multi-phase flow of bubbly liquids, fluid suspensions of particulates, and binary mixtures of simple fluids [3]. Other examples in condensed matter physics include two-fluid models for superfluid helium [4], dynamics of plasmas [5], transport in superconductors [6], viscoelasticity of concentrated fluid emulsions [7], flow-induced ordering of wormlike micelle solutions [8], flow of colloidal suspensions [9]. Two-fluid models have been used extensively to model a wide range of dynamical phenomena in polymer solutions and binary blends, including the hydrodynamics modes of quiescent polymer solutions [10, 11], kinetics of polymer dissolution [12], hydrodynamics and rheology of polymer solutions and blends [13]-[19], and polymer migration and phase separation under flow [20]-[27].

These examples share certain general features. In each, two distinct species or coexisting phases (gas and liquid, normal fluid and superfluid, polymer and solvent, mesogens and solvent etc.) with mass densities ρ_1 and ρ_2, which are conserved individually in the absence of chemical reactions, move with distinct velocities $\boldsymbol{v}_1$ and $\boldsymbol{v}_2$, respectively. Due to (usually strong) internal friction, the momenta of the constituent species, $\rho_1\boldsymbol{v}_1$ and $\rho_2\boldsymbol{v}_2$, are not conserved individually. Of course, total momentum is conserved. In most

cases of fluid mixtures the friction is so strong that the velocity difference $\boldsymbol{v}_1 - \boldsymbol{v}_2$ is nonzero for very short times only, i.e. it is a very rapidly relaxing quantity that is not included in the hydrodynamic description for binary mixtures. However, there are systems and situations, where the relaxation of the relative momenta is slow enough to have a significant influence even on the hydrodynamic time scale. Then a two-fluid description is appropriate and useful.

In this communication we focus on a general nonlinear two-fluid description of complex fluids, where one species is a viscous Newtonian fluid and the other either a polymer or a liquid crystal. Emphasis is placed on the rigorous derivation of the dynamic equations within the framework of hydrodynamics as contrasted to ad-hoc treatments. The resulting equations are rather general and complicated. They can and have to be simplified for special applications or systems by appropriate and well-defined approximations. One of the advantages of starting from the general theory is the possibility to identify and characterize the approximations made. The hydrodynamic method, described in some detail in [28]-[30], is quite general and rigorous, being based on symmetries, conservation laws, and thermodynamics. In the following sections, we provide a detailed analysis of two-fluid models for lyotropic nematogens in a simple viscous solvent, followed by an abbreviated extension of this treatment for isotropic elastomers (e.g. entangled polymer solutions and gels) in a simple viscous solvent. We close with a discussion of our general results and their possible implications for experiments.

2 Thermodynamics

The hydrodynamics of fluid mixtures as described above is governed by conservation laws (individual masses, total momentum and total energy), balance equations for the liquid crystalline degrees of freedom, for the transient elasticity of polymers and for the relaxation of relative momentum. There are different ways of writing the appropriate equations. One popular choice is to use equations for individual mass densities and individual momentum densities, another to use the mass density and one concentration variable and the total momentum density and the relative velocity difference. Since they both have their advantages and disadvantages we will present both ways of description and show, how they are connected. In this and the following sections we will use a nematic liquid crystal as the second, complex fluid. Transcription of the formulas to the polymer case will be given in Sec.(8).

The starting point of any macroscopic description is the total energy E of the system as a function of all the relevant variables. Since the energy is a first order Eulerian form of the extensive quantities, we can write

$$E = \epsilon V = \int \epsilon \, dV = E(M_1, \, M_2, \, V, \, \boldsymbol{G_1}, \, \boldsymbol{G_2}, \, S, \, M_2 \nabla_j n_i, \, M_2 \delta n_i) \qquad (1)$$

The masses, M_1, M_2 and momenta $\boldsymbol{G_1}$, $\boldsymbol{G_2}$ of species 1 and 2 are related to the appropriate

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(volume) densities by $\rho_1 = M_1/V$, $\rho_2 = M_2/V$, $\boldsymbol{g}_1 = \boldsymbol{G}_1/V = \rho_1\boldsymbol{v}_1$, $\boldsymbol{g}_2 = \boldsymbol{G}_2/V = \rho_2\boldsymbol{v}_2$, while for the entropy density $\sigma = S/V$. The nematic degrees of freedom are related to species 2 and consist of director rotations δn_i. The nematic phase shows orientational order along the line denoted by $\boldsymbol{n}$ (with $\boldsymbol{n}^2 = 1$) called the director. Since up and down (along that line) cannot be discriminated, all equations have to be invariant under a $\boldsymbol{n} \to -\boldsymbol{n}$ transformation. Homogeneous rotations do not cost energy, so in a linear description (of the field-free case) δn_i is absent in E and only gradients $\nabla_j n_i$ enter [31]. We have kept both terms to cope with the general case.

Introducing thermodynamic derivatives (partial derivatives where all other variables are kept fixed) we define temperature T, thermodynamic pressure p, chemical potentials μ_1, μ_2 and velocities $\boldsymbol{v}_1$, $\boldsymbol{v}_2$ of the two fluids, as well as the conjugate fields χ_{ij} and k_i connected to the nematic degrees of freedom

$$T = \frac{\partial E}{\partial S} = \frac{\partial \epsilon}{\partial \sigma}, \qquad \mu_1 = \frac{\partial E}{\partial M_1} = \frac{\partial \epsilon}{\partial \rho_1}, \qquad \mu_2 = \frac{\partial E}{\partial M_2} = \frac{\partial \epsilon}{\partial \rho_2},$$

$$p = -\frac{\partial E}{\partial V}, \qquad \boldsymbol{v}_1 = \frac{\partial E}{\partial \boldsymbol{G}_1} = \frac{\partial \epsilon}{\partial \boldsymbol{g}_1}, \qquad \boldsymbol{v}_2 = \frac{\partial E}{\partial \boldsymbol{G}_2} = \frac{\partial \epsilon}{\partial \boldsymbol{g}_2} \tag{2}$$

$$k_i = \frac{\partial E}{\partial (M_2 n_i)} = \frac{\partial \epsilon}{\partial (\rho_2 n_i)}, \qquad \chi_{ij} = \frac{\partial E}{\partial (M_2 \nabla_j n_i)} = \frac{\partial \epsilon}{\partial (\rho_2 \nabla_j n_i)}$$

Expanding eq.(1) into first order differentials, the condition $dV = 0$ leads to an expression for the pressure

$$p = -\epsilon + T\sigma + \rho_1 \mu_1 + \rho_2 \bar{\mu}_2 + \boldsymbol{v}_1 \cdot \boldsymbol{g}_1 + \boldsymbol{v}_2 \cdot \boldsymbol{g}_2 \tag{3}$$

where we have introduced the effective chemical potential of the nematic $\bar{\mu}_2 = \mu_2 + \chi_{ij}\nabla_j n_i + k_i \delta n_i$. In addition, the differentials are related by the Gibbs relation

$$d\epsilon = T d\sigma + \mu_1 \, d\rho_1 + \bar{\mu}_2 \, d\rho_2 + \boldsymbol{v}_1 \cdot d\boldsymbol{g}_1 + \boldsymbol{v}_2 \cdot d\boldsymbol{g}_2 + \Psi_{ij} \, d\nabla_j n_i + h_i \, dn_i \tag{4}$$

with the more familiar nematic conjugate fields $\Psi_{ij} = \rho_2 \chi_{ij}$ and $h_i = \rho_2 k_i$. From eqs.(3, 4) the expression for the differential pressure results (Gibbs-Duhem relation) that is useful in switching from pressure to chemical potentials or vice versa

$$dp = \sigma \, dT + \rho_1 \, d\mu_1 + \rho_2 \, d\bar{\mu}_2 + \boldsymbol{g}_1 \cdot d\boldsymbol{v}_1 + \boldsymbol{g}_2 \cdot d\boldsymbol{v}_2 - \Psi_{ij} \, d\nabla_j n_i - h_i \, dn_i \tag{5}$$

A second set of equations is obtained by switching to the total density, $\rho = \rho_1 + \rho_2$, and the total momentum, $\boldsymbol{g} = \boldsymbol{g}_1 + \boldsymbol{g}_2 = \rho_1\boldsymbol{v}_1 + \rho_2\boldsymbol{v}_2$, which are the sums of the original quantities and which are both conserved quantities. The two-fluid nature has then to be represented by additional variables. A natural choice seems to be the use of the density and momentum differences. However the latter choice is problematic, since it necessarily implies the conjugate quantities also to be the (arithmetic) sums and differences of the original conjugate quantities. Thus, the conjugate to $\boldsymbol{g}$ would be $\boldsymbol{v_1} + \boldsymbol{v_2}$, which does not reflect correctly the possible one-fluid limits $\rho_1 \to 0$ or $\rho_2 \to 0$. The physically acceptable conjugate to the total momentum is the mean velocity $\boldsymbol{v}$ defined by $\rho^{-1}\boldsymbol{g}$.

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Insisting on $\boldsymbol{v}$, the mean velocity, to be the conjugate of the total momentum $\boldsymbol{g}$, the choice of the remaining variable describing the different velocities is severely limited. Compatibility with (4) allows as variable only the velocity difference *** $\boldsymbol{w} \equiv \boldsymbol{v_1} - \boldsymbol{v_2}$ (with $\boldsymbol{m} \equiv \rho^{-1}\rho_1\rho_2\boldsymbol{w}$ as conjugate quantity) or more generally $\alpha\boldsymbol{w}$ as variable with $\alpha^{-1}\rho_1\rho_2\rho^{-1}\boldsymbol{w}$ as conjugate, where α can be freely choosen. There is no a-priori advantage for any of the choices and we will stick to $\alpha = 1$.[1] *** From $\boldsymbol{w} = \boldsymbol{g}_1/\rho_1 - \boldsymbol{g}_2/\rho_2$ one gets

$$\boldsymbol{v}_1 = \rho^{-1}\boldsymbol{g} + (1 - \phi)\boldsymbol{w}, \qquad \boldsymbol{v}_2 = \rho^{-1}\boldsymbol{g} - \phi\,\boldsymbol{w} \tag{6}$$

The representation of the two different densities is less problematic. A convenient choice for that variable is the concentration, $\phi = \rho_1/\rho$, with $\rho_2/\rho = 1 - \phi$. If the expansion coefficients of the two fluids are the same, ϕ can be interpreted as the volume fraction as well. Instead of ϕ one could have used, e.g. the density difference $\rho_1 - \rho_2$ (or any other linear combination of ρ_1 and ρ_2 different from ρ) as variable without much change.

After some trivial algebra eqs.(3-5) can be written in the new variables as

$$
\begin{align}
p &= -\epsilon + T\sigma + \rho\mu + \rho^{-1}\boldsymbol{g}^2 \tag{7} \\
d\epsilon &= T\,d\sigma + \Pi'\,d\phi + \mu\,d\rho + \boldsymbol{v} \cdot d\boldsymbol{g} + \boldsymbol{m} \cdot d\boldsymbol{w} + \Psi_{ij}\,d\nabla_j n_i + h_i\,dn_i \tag{8} \\
dp &= \sigma\,dT + \rho\,d\mu + \boldsymbol{g} \cdot d\boldsymbol{v} - \boldsymbol{m} \cdot d\boldsymbol{w} - \Pi'\,d\phi - \Psi_{ij}\,d\nabla_j n_i - h_i\,dn_i \tag{9}
\end{align}
$$

where we have introduced the relative pressure Π', the total chemical potential μ, the mean velocity $\boldsymbol{v}$ and the weighted relative momentum $\boldsymbol{m}$ defined by

$$
\begin{align}
\Pi' &= \rho\,(\mu_1 - \bar{\mu}_2) + \boldsymbol{w} \cdot \boldsymbol{g} + \rho\,\boldsymbol{w}^2(1 - 2\phi) \equiv \rho\,\Pi \\
\mu &= \mu_1\phi + \bar{\mu}_2(1 - \phi) + \boldsymbol{w}^2\phi(1 - \phi) \\
\text{or vice versa} \qquad \mu_1 &= \mu + \rho^{-1}\rho_2\,(\Pi - \boldsymbol{w} \cdot \boldsymbol{v}_1) \tag{10} \\
\mu_2 &= \mu - \rho^{-1}\rho_1\,(\Pi + \boldsymbol{w} \cdot \boldsymbol{v}_2) \\
\text{where} \qquad \boldsymbol{v} &= \phi\,\boldsymbol{v}_1 + (1 - \phi)\,\boldsymbol{v}_2 = \rho^{-1}(\boldsymbol{g_1} + \boldsymbol{g_2}) \\
\boldsymbol{m} &= \rho\,(1 - \phi)\phi\,\boldsymbol{w} = (\rho_2\boldsymbol{g}_1 - \rho_1\boldsymbol{g}_2)\rho^{-1}
\end{align}
$$

The Gibbs relations connects variables that show different rotational behavior. Energy, entropy, the densities and the concentration are scalar quantities that do not change under (rigid) rotations, i.e. $d\epsilon = d\sigma = d\rho = d\rho_1 = d\rho_2 = d\phi = 0$. The vectors and tensors are transformed according to $dn_i = \Omega_{ij}n_j$, $dg_i = \Omega_{ij}g_j$, $dw_i = \Omega_{ij}w_j$, $d\nabla_j n_i = \Omega_{jk}\nabla_k n_i + \nabla_j\Omega_{ik}n_k$, where $\Omega_{ij} = -\Omega_{ji}$ is any constant antisymmetric matrix. The rotational invariance of the Gibbs relation (4,8) then leads to the relation

$$h_i n_j + \Psi_{ki}\nabla_j n_k + \Psi_{ik}\nabla_k n_j = h_j n_i + \Psi_{kj}\nabla_i n_k + \Psi_{jk}\nabla_k n_i \tag{11}$$

which has to be fulfilled by the conjugate quantities. There are no contributions from the momenta and velocities, since $\boldsymbol{g} \parallel \boldsymbol{v}$, $\boldsymbol{w} \parallel \boldsymbol{m}$, and $\boldsymbol{g_{1,2}} \parallel \boldsymbol{v_{1,2}}$. Relation (11) is useful for reformulating the stress tensor, in particular to symmetrize it explicitly [32].

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[1]The choice $\alpha = \rho_1\rho_2\rho^{-1}$ would just interchange the roles of $\boldsymbol{w}$ and $\boldsymbol{m}$ as variable and conjugate.

Having set up the thermodynamics of the relevant variables we are now in a position to establish the structure of the dynamic equations.

3 Dynamic Equations

For the two fluids there are independent continuity equations stating that neither mass can be destroyed nor created, but only transported. Transport can involve convection as well as (relative) diffusion. This leads immediately to

$$\dot{\rho}_1 + \nabla_i(\rho_1 v_i^{(1)} + j_i^{(1)}) = 0 \tag{12}$$

$$\dot{\rho}_2 + \nabla_i(\rho_2 v_i^{(2)} - j_i^{(1)}) = 0 \tag{13}$$

When dealing with components of vectors, the subscripts 1, 2 are written as superscripts for clarity. The phenomenological mass currents in Eqs.(12, 13) add up to zero, since the total mass current is equal to the total momentum density $\boldsymbol{g} = \rho\,\boldsymbol{v}$. Eqs.(12, 13) can be rewritten in terms of the total density and the concentration as

$$\dot{\rho} + \nabla_j \rho\, v_j = 0 \tag{14}$$

$$\dot{\phi} + v_j \nabla_j \phi + \rho^{-1} \nabla_i \left(\rho\phi(1-\phi)w_i + j_i^{(1)} \right) = 0 \tag{15}$$

show the characteristic difference between extensive quantities, where convection is of the form $\boldsymbol{\nabla} \cdot (\boldsymbol{v}*)$ and intensive ones with $\boldsymbol{v} \cdot \boldsymbol{\nabla}*$.

Note that the concentration does not obey a conservation law, except when linearized around a zero-velocity state or if $\rho = const.$ is assumed. Because the mass current density of the total fluid is equal to the momentum density $\boldsymbol{g}$ $(= \rho\boldsymbol{v})$, the total mass is convected by the mean velocity in (14). In Eqs. (12, 13, 15) the convective terms are not fixed a priori, since the phenomenological current $\boldsymbol{j}_1^{(1)}$ can contain contributions proportional to some velocities, thus altering the effective velocity, with which the different quantities are convected. We will discuss this point extensively after having derived the full set of equations.

The dynamic equations for the other variables are even more complicated and also contain phenomenological parts. These are expressed by yet to be determined currents. But they also contain convective (or transport) terms. Therefore, we can set up the following equations as an ansatz

$$\dot{\epsilon} + \nabla_j(\epsilon + p)v_j + \nabla_i j_i^{(\epsilon)} = 0 \tag{16}$$

$$\dot{\sigma} + \nabla_j \sigma v_j + \nabla_i j_i^{(\sigma)} = R/T \tag{17}$$

$$\dot{g}_i + \nabla_j g_i v_j + \nabla_j \sigma_{ij} = 0 \tag{18}$$

$$\dot{w}_i + v_j \nabla_j w_i + X_i = 0 \tag{19}$$

$$\dot{n}_i + v_j \nabla_j n_i + Y_i = 0 \tag{20}$$

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containing either the divergence of a current ($j_i^{(\epsilon)}$, $j_i^{(\sigma)}$, $j_i^{(1)}$, σ_{ij}) when conservation laws are involved, or quasi-currents (X_i, Y_i) in the case of balance equations for non-conserved variables. Each of the currents and quasi-currents consists generally of three parts: A geometric or symmetry related one without any phenomenological coefficients, which we will determine below, and two phenomenological parts, which are either reversible (superscript rev) or irreversible (superscript dis). The phenomenological parts will be discussed in section 5. The entropy balance (17) is not a conservation law, since for irreversible processes the entropy production R has to be positive and only for purely reversible actions $R = 0$. In Eqs. (14– 20) the convective terms are written down such that all quantities are convected by the *same* velocity.[2] This is dictated by the postulation of zero entropy production (these transport terms are reversible). However, it should be repeated that the phenomenological reversible currents may change the effective convection velocity, something we will discuss later.

Putting the dynamic equations (14–20) into the Gibbs relation (4) the condition $R = 0$ ($R > 0$) for the convective and the reversible (dissipative) phenomenological parts of the currents, leads to the following conditions

$$
\begin{align}
\sigma_{ij} &= \delta_{ij}p + \Psi_{kj}\nabla_i n_k + \sigma_{ij}^{(rev)} + \sigma_{ij}^{(dis)} \tag{21}\\
X_i &= \nabla_i\Pi + X_i^{(rev)} + X_i^{(dis)} \tag{22}\\
Y_i &= Y_i^{(rev)} + Y_k^{(dis)} \tag{23}\\
j_i^{(\sigma)} &= j_i^{(\sigma,rev)} + j_i^{(\sigma,dis)} \tag{24}\\
j_i^{(1)} &= j_i^{(1,rev)} + j_i^{(1,dis)} \tag{25}
\end{align}
$$

with the generalized conjugate to the nematic degrees of freedom $\bar{h}_i = h_i - \nabla_j\Psi_{ij} = \rho_2(k_i - \nabla_j\chi_{ij})$. The stress tensor σ_{ij} contains the isotropic pressure p (7), while the quasi-current X_i of the relative velocity contains the gradient of Π, the relative pressure divided by the total density, (10). The terms related to the nematic degrees of freedom are well-known from ordinary nematodynamics. The energy conservation law is redundant here, because of the Gibbs relation (4) and $j_i^{(\epsilon)}$ is not needed.

The phenomenological parts have to fulfill (up to an irrelevant divergence term)[3]

$$
R = -j_i^{(\sigma,*)}\nabla_i T - j_i^{(1,*)}\nabla_i\Pi - \sigma_{ij}^{(*)}\nabla_j v_i + \bar{h}_i Y_i^{(*)} + m_i X_i^{(*)} \geq 0 \tag{26}
$$

with the equal sign ($>$ sign) for $* = rev$ ($* = dis$), respectively.

Eq.(26) also reveals the equilibrium conditions

$$
\nabla_i T = 0 \qquad \nabla_i\Pi = 0 \qquad A_{ij} = 0 \qquad \bar{h}_i = 0 \qquad m_i = 0 \tag{27}
$$

where $2A_{ij} = \nabla_j v_i + \nabla_i v_j$.

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[2]In the energy conservation law (16) the free enthalpy $\epsilon + p$ is convected, cf.[30]

[3]The true condition is $\int R\, dV \geq 0$.

Before we will determine the phenomenological parts in (21–24), we first have a look into the 2-fluid statics.

4   Statics

The statics is given by the connection of thermodynamic conjugates with the variables. The conjugates are defined by partial derivatives of the energy density (2). Thus one can either write down a phenomenological energy expression and take the derivatives or give directly these relations under the proviso that mixed derivatives are equal. Two of these connections have already been given in eq.(10) relating $\boldsymbol{v}$ with $\boldsymbol{g}$ and $\boldsymbol{m}$ with $\boldsymbol{w}$. Of course, these are not really static relations. They are fixed (and not of phenomenological nature), since the mass current $\rho\boldsymbol{v}$ is identical to the momentum density and since the kinetic energy density is $(1/2)\rho_1\boldsymbol{v}_1^2 + (1/2)\rho_2\boldsymbol{v}_2^2$.

The 3 scalar conjugates $\{T, \Pi, \mu\}$ have to be expressed by the variables $\{\sigma, \phi, \rho\}$ or using the other set of variables $\{\sigma, \rho_1, \rho_2\}$ and conjugates $\{T, \mu_1, \mu_2\}$ by

$$\delta T = \frac{T}{C_V}\delta\sigma + \frac{1}{\rho\alpha_1}\delta\rho_1 + \frac{1}{\rho\alpha_2}\delta\rho_2 \tag{28}$$

$$\mu_1 = \frac{1}{\rho^2\kappa_1}\delta\rho_1 + \frac{1}{\rho^2\kappa_3}\delta\rho_2 + \frac{1}{\rho\alpha_1}\delta\sigma \tag{29}$$

$$\bar{\mu}_2 = \frac{1}{\rho^2\kappa_2}\delta\rho_2 + \frac{1}{\rho^2\kappa_3}\delta\rho_1 + \frac{1}{\rho\alpha_2}\delta\sigma \tag{30}$$

The other conjugates Π and μ have been related to μ_1 and $\bar{\mu}_2$ in (10) and are therefore also fully determined

$$\delta T = \frac{T}{C_V}\delta\sigma + \frac{1}{\alpha_\phi}\delta\phi + \frac{1}{\rho\alpha_\rho}\delta\rho \tag{31}$$

$$\Pi = \frac{1}{\rho\kappa_\phi}\delta\phi + \frac{1}{\rho^2\kappa_\pi}\delta\rho + \frac{1}{\rho\alpha_\phi}\delta\sigma + \boldsymbol{w}\cdot\boldsymbol{v} + \boldsymbol{w}^2(1 - 2\phi) \tag{32}$$

$$\mu = \frac{1}{\rho^2\kappa_\mu}\delta\rho + \frac{1}{\rho\kappa_\pi}\delta\phi + \frac{1}{\rho\alpha_\rho}\delta\sigma + \boldsymbol{w}^2\phi(1 - \phi) \tag{33}$$

with

$$\alpha_\phi{}^{-1} = \alpha_1{}^{-1} - \alpha_2{}^{-1} \tag{34}$$

$$\alpha_\rho{}^{-1} = \phi\alpha_1{}^{-1} + (1 - \phi)\alpha_2{}^{-1} \tag{35}$$

$$\kappa_\phi{}^{-1} = \kappa_1{}^{-1} + \kappa_2{}^{-1} - 2\kappa_3{}^{-1} \tag{36}$$

$$\kappa_\pi{}^{-1} = \phi\kappa_\phi{}^{-1} - \kappa_2{}^{-1} + \kappa_3{}^{-1} \tag{37}$$

$$\kappa_\mu{}^{-1} = \phi^2\kappa_1{}^{-1} - (1 - \phi)^2\kappa_2{}^{-1} \tag{38}$$

Eqs.(28–30) as well as (31–33) contain 6 static susceptibilities as compared to 3 in a 1-fluid description. In addition to the specific heat C_V there are 2 thermal expansion coefficients (since there are 2 densities) and 3 compressibilities (2 diagonal and one cross term). Eqs.(28–30) are linear in the deviations from equilibrium, while (31–33) explicitly contains nonlinear corrections involving velocities. Of course, the coefficients can still be phenomenological functions of the scalar variables (e.g. T or σ, p or ρ, ρ_1 and ρ_2 and even $\boldsymbol{w}^2$) giving rise to additional nonlinearities that come with (usually) small coefficients. Note that neglecting cross-susceptibilities either in (28–30) or in (31–33) denotes two physically distinct (and incompatible) approximations, the justification of either one is not obvious a priori.

Of course, there are situations where one has to go beyond the approximation used in the static equations above. Describing spinodal decomposition of the fluids, e.g. by an energy density $\epsilon = a\,\phi^2 + b\,[(1-\phi)\ln(1-\phi) + \chi\phi(1-\phi)] + c\,\phi^4 + d\,(\nabla_1\phi)^2$ immediately leads to nonlinear and gradient terms w.r.t. ϕ.

What is left is the determination of $\bar{h}_i = h_i - \nabla_j\Psi_{ij}$ in terms of n_i; cross-couplings to other variables are not possible due to symmetry. Thus this part of the statics is identical to that of ordinary nematics and can be taken over without any change

$$\Psi_{ij} = K_{ijkl}\nabla_l n_k \tag{39}$$

$$h_i = \delta_{iq}^{\perp}\frac{\partial K_{pjkl}}{2\partial n_q}(\nabla_j n_p)(\nabla_l n_k) - \chi_a(\boldsymbol{H}\!\cdot\!\boldsymbol{n})H_i - \epsilon_a(\boldsymbol{E}\!\cdot\!\boldsymbol{n})E_i \tag{40}$$

with $K_{ijkl} = K_1\,\delta_{ij}^{\perp}\delta_{kl}^{\perp} + K_2\,n_p\,\epsilon_{pij}\,n_q\,\epsilon_{qkl} + K_3\,n_j\,n_l\,\delta_{ik}^{\perp}$, the Frank gradient energy, and the transverse Kronecker symbol, $\delta_{ij}^{\perp} = \delta_{ij} - n_i n_j$. Orientation effects due to static external magnetic and electric effects enter through the diamagnetic (χ_a) and dielectric (ϵ_a) anisotropy. For positive anisotropies the director is parallel to the external magnetic or electric field in equilibrium, which leads to a restoring torque outside equilibrium, e.g. to a (linearized) contribution to $h_i = \chi_a\boldsymbol{H}^2\delta n_i$ (with $n_i\delta n_i = 0$). For negative anisotropies the director is perpendicular to the external fields and e.g. $h_i = |\chi_a|(\boldsymbol{H}\cdot\delta\boldsymbol{n})H_i$.

Since Ψ_{ij} and h_i are proportional to ρ_2, so are the K_n's (and χ_a, ϵ_a). Again (39) is linear in the deviations from equilibrium, but the inherent dependence of the material tensor on the direction $\boldsymbol{n}$ leads to nonlinearities in (40).

5 Phenomenological Part of the Dynamics

We now close our system of equations by setting up the connection between the currents and the thermodynamic conjugates (or rather their gradients usually called thermodynamic forces). For the irreversible part this is done by writing the entropy production in

terms of the forces

$$2R = \kappa_{ij}(\nabla_i T)(\nabla_j T) + D_{ij}(\nabla_i \Pi)(\nabla_j \Pi) + 2D_{ij}^{(T)}(\nabla_i \Pi)(\nabla_j T) + \gamma_1^{-1}\delta_{ij}^\perp \bar{h}_i \bar{h}_j$$
$$+ \nu_{ijkl}(\nabla_j v_i)(\nabla_l v_k) + \xi'_{ij} m_i m_j \tag{41}$$

including heat conduction, diffusion and thermodiffusion (κ_{ij}, D_{ij}, $D_{ij}^{(T)}$, respectively, all of the form $\kappa_{ij} = \kappa_\perp \delta_{ij}^\perp + \kappa_\parallel n_i n_j$), director orientational viscosity γ_1, and viscosity related to gradients of the mean velocity ν_{ijkl}. The latter has a $\nu_{ijkl} = \nu_{klij}$ symmetry and is of the uniaxial form [28] characteristic for nematic systems. The last term in (41) describes the mutual friction between the two species as will become clear below. In (41 we have neglected viscosity-like contributions involving the relative velocity $\nabla_j m_i$, since there is already dissipation due to m_i. A more complete discussion of viscosity in a 2-fluid discussion is given in the Appendix. The dissipation function given above is bilinear in the forces, an approximation commonly called linear irreversible thermodynamics. Nevertheless it leads to nonlinearities due to (implicit and explicit) dependences of transport tensors on the variables.

According to (26) the dissipative parts of the phenomenological currents then follow from differentiating R

$$j_i^{(\sigma,dis)} = -(\partial R)/(\partial \nabla_i T) = -\kappa_{ij}\nabla_j T - \rho\,\phi(1-\phi)\,d_{ij}^{(T)}\nabla_j \Pi \tag{42}$$

$$Y_i^{(dis)} = (\partial R)/(\partial \bar{h}_i) = \gamma_1^{-1}\delta_{ij}^\perp \bar{h}_j \tag{43}$$

$$\sigma_{ij}^{(dis)} = -(\partial R)/(\partial \nabla_j v_i) = -\nu_{ijkl}\,\nabla_l v_k \tag{44}$$

$$X_i^{(dis)} = (\partial R)/(\partial m_i) = \xi'_{ij}\,m_j \tag{45}$$

$$j_i^{(1,dis)} = -(\partial R)/(\partial \nabla_i \Pi) = -\rho\,d_{ij}\nabla_j \Pi - \rho\,\phi(1-\phi)\,d_{ij}^{(T)}\nabla_j T \tag{46}$$

where we have introduced the usual form of the diffusion ($D_{ij} = \rho\,d_{ij}$) as well as the thermo-diffusion tensor ($D_{ij}^{(T)} = \rho\,\phi(1-\phi)\,d_{ij}^{(T)}$). The ratios $d_*^{(T)}/d_*$ (with $* \in \{\perp, \parallel\}$) and $d_*^{(T)}/\kappa_*$ are called the Soret and the Dufour coefficients, respectively (the latter being neglected usually in liquids). The viscosity term in (44) has the same form as in a 1-fluid description. For a more general treatment of viscosity-like contributions cf. Appendix.

In ad-hoc treatments of 2-fluid systems the mutual friction of the two species is introduced via an interaction force $\boldsymbol{f}_{12}$ in the momentum equations for the single fluids, $\rho_1 \dot{\boldsymbol{v}}_1 = \boldsymbol{f}_{12}$ and $\rho_2 \dot{\boldsymbol{v}}_2 = -\boldsymbol{f}_{12}$ preserving total momentum. The force is related to the velocity difference, $\boldsymbol{f}_{12} = -\xi\,\rho_1\,\rho_2\,\boldsymbol{w}$ and is non-zero only if both fluids are present. This translates directly into $\dot{\boldsymbol{w}} = -\xi\,\rho\,\boldsymbol{w}$ and can be compared to (45). First, in a nematic environment the force $\boldsymbol{f}_{12}$ is not necessarily parallel to $\boldsymbol{w}$ due to the possible anisotropy, rendering the ξ to be a tensor $\xi_{ij} = \xi_\perp \delta_{ij}^\perp + \xi_\parallel n_i n_j$. Then comparison with (45) gives $\xi_{ij} = \phi(1-\phi)\xi'_{ij}$, which shows that the ad-hoc choice for $\boldsymbol{f}_{12}$ is the only possible one within linear irreversible thermodynamics. Of course, there is room for suitable nonlinear extensions (e.g. $\sim \boldsymbol{f}_{12}^3$ or ξ being a function of scalar state variables like T, ϕ, or ρ etc.).

The reversible part of the dynamics is either dictated by symmetries or phenomenological. The symmetry parts have been discussed in sec.5 and are listed in eqs.(21–24). The

phenomenological reversible currents *cannot* be derived from any potential (especially not from any kind of Hamiltonian, despite being reversible). They are most easily derived by writing down all symmetry-allowed contributions to the various currents and then make sure that the entropy production (26) is zero. We find

$$
\begin{align}
Y_i^{(rev)} &= -\lambda_{ijk}\nabla_j v_k - \lambda_{ijk}^{(m)}\nabla_j m_k + \beta_1 m_j \nabla_j n_i \tag{47}\\
\sigma_{ij}^{(rev)} &= -\lambda_{kji}\bar{h}_k + 2\beta_2\, m_i\, w_j + \beta_2'(m_i g_j + m_j g_i) \tag{48}\\
X_i^{(rev)} &= \beta_{ij}\,\nabla_j T + \gamma_{ij}\,\nabla_j\Pi - \nabla_j(\lambda_{kji}^{(m)}\,\bar{h}_k) - \beta_1\,\bar{h}_j\,\nabla_i n_j \\
&\quad + (\beta_2\,w_j + \beta_2'\,g_j)(\nabla_j v_i + \nabla_i v_j) + \beta_3 m_j(\nabla_j w_i - \nabla_i w_j) \\
&\quad + \beta_4 w_j(\nabla_j v_i - \nabla_i v_j) + \beta_5(m_i m_j\nabla_j - \boldsymbol{m}^2\nabla_i)F \tag{49}\\
j_i^{(\sigma,rev)} &= \beta_{ij}\,m_j \tag{50}\\
j_i^{(1,rev)} &= \gamma_{ij}\,m_j \tag{51}
\end{align}
$$

with $2\lambda_{ijk} = \lambda_1\delta_{ij}^\perp n_k + \lambda_2\delta_{ik}^\perp n_j$, $2\lambda_{ijk}^{(m)} = \lambda_1^{(m)}\delta_{ij}^\perp n_k + \lambda_2^{(m)}\delta_{ik}^\perp n_j$, and $\beta_{ij} = \beta_\perp\delta_{ij}^\perp + \beta_\parallel n_i n_j$ and $\gamma_{ij} = \gamma_\perp\delta_{ij}^\perp + \gamma_\parallel n_i n_j$ and F any function of the scalar variables or conjugates (e.g. T, ρ, ϕ). Since the term involving F is already of cubic order, we will neglect it in the following and suppress similar terms in other equations. Of the four flow alignment parameters λ, only three are independent as will be discussed below. The β_{ij}-tensor in (49,50) describes a reversible entropy (energy) current due to a non-zero velocity difference as well as a change in the velocity difference due to a temperature gradient. In the limit of large ξ the γ and β parameters are related to diffusion and thermodiffusion (see below). The 1-fluid description is obtained in the limit of very large mutual friction, $\xi \to \infty$, which implies $\boldsymbol{w} \to 0$. The usual nematodynamics (with an additional concentration variable) is regained, while $\rho\xi\boldsymbol{w}$ stays finite accommodating Eqs.(19,45,49) and is slaved by the other variables.

6 Convective Velocities, Stress Division, and Concentration Dynamics

In (47–51) we have introduced terms, which are compatible with symmetries and $R = 0$, involving quadratic nonlinearities in the different velocities. Among them the β_1 term has a form quite similar to the convective term in (20). Thus the actual velocity, with which n_i is convected, is $\boldsymbol{v}_{conv} = \rho^{-1}\rho_1(\beta_1\rho_2 + 1)\boldsymbol{v}_1 + \rho^{-1}\rho_2(-\beta_1\rho_1 + 1)\boldsymbol{v}_2$ and can be either $\boldsymbol{v}_1$, $\boldsymbol{v}_2$ or something in-between, depending on β_1. Since it is hard to imagine that n_i is convected with a velocity larger than $max(|\boldsymbol{v}_1|, |\boldsymbol{v}_2|)$, β_1 is bounded $\rho_1^{-1} < \beta_1 < -\rho_2^{-1}$. If one accepts the reasonable assumption that n_i is convected with the velocity of the nematic fluid $\boldsymbol{v}_2$, then $\beta_1 = -\rho_2^{-1}$ is fixed (while for $\beta_1 = 0$, there is $\boldsymbol{v}_{conv} = \boldsymbol{v}$).

The choice of $\boldsymbol{v}_{conv}$ has additional implications for the flow alignment parameters λ. Since the director does not rotate in a frame that corotates with it, the quasicurrent Y_i couples to the vorticity by $Y_1^{rot} = \epsilon_{ijk}n_j\omega_k^{conv}$, where $2\omega_k^{conv} \equiv \text{curl}\,\boldsymbol{v}_{conv}$. For $\boldsymbol{v}_{conv} = \boldsymbol{v}$ this implies $\lambda_1^{(m)} = \lambda_2^{(m)}$ and $\lambda_2 - \lambda_1 = 2$, or in the usual parameterization $\lambda_1 = \lambda - 1$ and $\lambda_2 = \lambda + 1$. For $\boldsymbol{v}_{conv} = \boldsymbol{v}_2$ the conditions are

$$\lambda_1 = \lambda - 1, \qquad \lambda_2 = \lambda + 1, \qquad -\rho_2\lambda_1^{(m)} = \bar{\lambda} - 1, \qquad -\rho_2\lambda_2^{(m)} = \bar{\lambda} + 1 \qquad (52)$$

In any case only two of the λ's are independent.

The phenomenological contribution $\sim \gamma_{ij}$ in (51) affects the convection of the densities ρ_1 and ρ_2 in (12, 13). For $\gamma_\perp = \gamma_\parallel = 0$ the densities are convected with $\boldsymbol{v}_1$ and $\boldsymbol{v}_2$, respectively, while ϕ in (15) moves with the velocity $(1/\rho)(\rho_2\boldsymbol{v}_1 + \rho_1\boldsymbol{v}_2)$. All these quantities ($\rho_1$, ρ_2, ρ, and ϕ) are convected with $\boldsymbol{v}$ for $\gamma_\perp = \gamma_\parallel = -1$. Of course, other choices of the γ's (made either by a theorist or by nature!) will lead to different convection velocities.

Somewhat more involved is the question of the convective velocity for the different momenta. Because of $\boldsymbol{g} = \rho\boldsymbol{v}$, the total momentum has to be convected with $\boldsymbol{v}$ requiring $\beta_2' = 0$. If also $\boldsymbol{w}$ is convected with $\boldsymbol{v}$ then $\beta_2 = \beta_3 = \beta_4 = 0$, additionally, with the consequence that also the individual momenta, $\boldsymbol{g}_1$ and $\boldsymbol{g}_2$, are convected with $\boldsymbol{v}$. On the other hand, for $\boldsymbol{g}_1$ and $\boldsymbol{g}_2$ to be convected with $\boldsymbol{v}_1$ and $\boldsymbol{v}_2$, respectively, i.e.

$$\dot{g}_i^{(1)} + \nabla_j g_i^{(1)} v_j^{(1)} + X_i^{(1)} = 0 \qquad (53)$$
$$\dot{g}_i^{(2)} + \nabla_j g_i^{(2)} v_j^{(2)} + X_i^{(2)} = 0 \qquad (54)$$

where

$$X_i^{(1)} = \frac{\rho_1\rho_2}{\rho}X_i + \frac{\rho_1}{\rho}\nabla_j\sigma_{ij} - \frac{\rho_1}{\rho}w_i\nabla_j m_j - m_j\nabla_j v_i^{(1)} \qquad (55)$$
$$X_i^{(2)} = -\frac{\rho_1\rho_2}{\rho}X_i + \frac{\rho_2}{\rho}\nabla_j\sigma_{ij} - \frac{\rho_2}{\rho}w_i\nabla_j m_j + m_j\nabla_j v_i^{(2)} \qquad (56)$$

in order to be compatible with (18,19), the β-parameters have to be $\beta_2 = 1/2$, $\beta_3 = (1/\rho_1) - (1/\rho_2)$, and $\beta_4 = 1/2$, thus ensuring that X_i and X_2 do not contain additional transport terms. This choice of parameters results in $(\rho_2\boldsymbol{v}_1 + \rho_1\boldsymbol{v}_2)/\rho$ to be the convective velocity for $\boldsymbol{w}$ (which is the same as for ϕ, when $\rho_{1,2}$ are convected with $\boldsymbol{v}_{1,2}$). In addition the momentum current density due to flow then reads $g_j v_i + w_j m_i = \rho_1 v_i^{(1)} v_j^{(1)} + \rho_2 v_i^{(2)} v_j^{(2)}$, which is the expected expression.

The terms proportional to $\bar{h}_i$ in (48,49) constitute forces due to the nematic orientational elasticity. Generally they act on both fluids. Using (55,56) they read in linearized form

$$\dot{g}_i^{(1)}\big|_{nem} = \phi\left(\lambda_{kji} + \rho_2\lambda_{kji}^{(m)}\right)\nabla_j\bar{h}_k \qquad (57)$$
$$\dot{g}_i^{(2)}\big|_{nem} = (1 - \phi)\left(\lambda_{kji} - \rho_1\lambda_{kji}^{(m)}\right)\nabla_j\bar{h}_k \qquad (58)$$

Hence, for $\lambda_{ijk}^{(m)} = 0$ ($\lambda_1^{(m)} = 0 = \lambda_2^{(m)}$) this nematic force is distributed on fluid 1 and fluid 2 according to the ratio of ρ_1/ρ_2. It should be noted, however, that this kind of

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nematic stress division is only compatible with the choice of $\boldsymbol{n}$ being convected by $\boldsymbol{v}$, while it is incompatible with the choice of $\boldsymbol{v}_2$ as the convective velocity for $\boldsymbol{n}$ (cf. (52)). Another reasonable case for the stress division problem is obtained for $\lambda_{ijk} = -\rho_2 \lambda_{ijk}^{(m)}$ ($\lambda_1 = -\rho_2 \lambda_1^{(m)}$ and $\lambda_2 = -\rho_2 \lambda_2^{(m)}$). In that case the nematic force only acts on fluid 2 (the nematic component). This case is compatible with fluid 2 being convected with $\boldsymbol{v}_2$ and the force then reads

$$\dot{g}_i^{(2)}|_{nem} = \frac{1}{2}\left((\lambda-1)\delta_{kj}^{\perp}n_i + (\lambda+1)\delta_{ki}^{\perp}n_j\right)\nabla_j \bar{h}_k = \lambda_{kji}\nabla_j \bar{h}_k \tag{59}$$

which is the form familiar from 1-fluid nematics. Thus, the so-called stress division problem (how $\bar{h}_i$ in the stress tensor is divided between the two fluids) depends not only on specific material properties expressed by the phenomenological parameters $\lambda_{1,2}^{(m)}$ and $\lambda_{1,2}$, but is also intrinsically linked to the question of the appropriate convection velocity.

In order to verify experimentally the choices above, it seems to be difficult to directly measure specific convection velocities or the division of the nematic stress. However, there are situations, where these choices can be verified indirectly. Linearizing the dynamic equation for the relative velocity (19,22,45,49) and Fourier transform it w.r.t. time, $\boldsymbol{w}$ can be expressed by all the other variables. This can be used to eliminate $\boldsymbol{w}$ e.g. from the linearized dynamic equation for the concentration (15) leading to

$$i\omega\phi - d_{ij}^{eff}\nabla_i\nabla_j\Pi - \frac{\rho_1\rho_2}{\rho^2}d_{ij}^{(T)eff}\nabla_i\nabla_j T + \lambda^{(\phi)}(\boldsymbol{n}\cdot\boldsymbol{\nabla})div\boldsymbol{h} = 0 \tag{60}$$

where contributions of order $O(\nabla^4)$ have been neglected. The effective diffusion and thermo-diffusion (Soret) coefficients have got additional frequency dependent contributions due to the 2-fluid degree of freedom

$$d_*^{eff} = d_* + \frac{\rho_1\rho_2}{\rho^2}\frac{(1+\gamma_*)^2}{\rho\,\xi_* + i\omega} \tag{61}$$

$$d_*^{(T)eff} = d_*^{(T)} + \frac{\beta_*(1+\gamma_*)}{\rho\,\xi_* + i\omega} \tag{62}$$

where the subscript $_*$ stands for either $_\|$ or $_\perp$. There is also a dynamic coupling to the nematic degree of freedom due to

$$\lambda^{(\phi)} = \frac{\rho_1\rho_2}{2\rho^2}\left(\lambda_1^{(m)}\frac{1+\gamma_\|}{\rho\xi_\| + i\omega} + \lambda_2^{(m)}\frac{1+\gamma_\perp}{\rho\xi_\perp + i\omega}\right) \tag{63}$$

These possible additions to the concentration dynamics, however, depend on the choices for the convection velocities as well as on the way how the nematic stress has been divided among the two fluids. Assuming the densities $\rho_{1,2}$ to be convected with the mean velocity $\boldsymbol{v}$ (implying $\gamma_\| = \gamma_\perp = -1$) the additional contributions to diffusion and thermo-diffusion are all zero as well as the coupling to the nematic director. On the other hand, for $\rho_{1,2}$ to be convected with $\boldsymbol{v}_{1,2}$ respectively ($\gamma_\| = \gamma_\perp = 0$), both, diffusion and thermo-diffusion show

a dispersion step around $\omega \approx \rho\,\xi$. For a nematic stress division among fluid 1 and 2 according to the ratio ρ_1/ρ_2 (implying $\lambda_1^{(m)} = \lambda_2^{(m)} = 0$) there is no dynamic influence of the nematic degree of freedom on the concentration ($\lambda^{(\phi)} = 0$), while for any other choice there is one. In particular, if only fluid 2 carries nematic stress (and $\rho_{1,2}$ are convected with $\boldsymbol{v}_{1,2}$, respectively), this dynamic coupling is given by $\lambda^{(\phi)} = -(\rho_1/2\rho^3)[\bar{\lambda}(\xi_\parallel^{-1}+\xi_\perp^{-1})-\xi_\parallel^{-1}+\xi_\perp^{-1}]$ for strong friction ($\rho\xi_* \gg \omega$).

7 Simplified 2-Fluid Nematic Equations

In the preceding sections we have derived the most general and complete set of 2-fluid equations for a nematic and Newtonian mixture. Special emphasis has been laid on the correct form of the nonlinearities that come with the 2-fluid description. However, these equations are for most purposes unnecessarily complicated and can be simplified using reasonable assumptions. Starting from the correct general equations such assumptions, clearly spelled out, lead to controlled approximations and to a set of 2-fluid equations, whose limitations and implicit assumptions are clear and well defined in contrast to most ad-hoc approaches.

Here we want to display explicitly 2-fluid hydrodynamics for a nematic/simple fluid mixture under the following assumptions,
a) convection with natural velocities (for $\boldsymbol{n}$, $\boldsymbol{g}_2$, ρ_2 and $\boldsymbol{g}_1$, ρ_1 this is $\boldsymbol{v}_2$ and $\boldsymbol{v}_1$, respectively, or explicitly $\beta_1 = -\rho_2^{-1}$, $\beta_2 = \beta_4 = 1/2$, $\beta_3 = \rho_1^{-1} - \rho_2^{-1}$, $\gamma_\perp = 0 = \gamma_\parallel$ and eq.(52));
b) the linearized orientation-elastic force acts on the nematic fluid (index 2) only (i.e. $\lambda_{1,2} = -\rho_2\lambda_{1,2}^{(m)}$);
c) global incompressibility, $\delta\rho = 0$ (i.e. $\delta\rho_1 = -\delta\rho_2$);
d) neglecting the phenomenological reactive entropy current ($\beta_{ij} = 0$);
e) linearizing the phenomenological dissipative currents, but keeping quadratic nonlinearities otherwise.

Then the following set of equations is obtained:
The incompressibility condition

$$0 = \operatorname{div}\boldsymbol{v} \tag{64}$$

$$\text{or}\quad 0 = \boldsymbol{w}\cdot\boldsymbol{\nabla}\rho_1 + \rho_1\operatorname{div}\boldsymbol{v}_1 + \rho_2\operatorname{div}\boldsymbol{v}_2 \tag{65}$$

$$\text{or}\quad 0 = \boldsymbol{w}\cdot\boldsymbol{\nabla}\phi + \phi\operatorname{div}(1-\phi)\boldsymbol{w} - (1-\phi)\operatorname{div}\phi\boldsymbol{w} \tag{66}$$

the concentration dynamics

$$\dot{\phi} + \nabla_i\left(\phi v_i + \phi(1-\phi)w_i\right) - d_{ij}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) - \phi(1-\phi)d_{ij}^{(T)}\nabla_j\nabla_i T = 0 \tag{67}$$

$$\text{or}\quad \dot{\rho}_1 + \boldsymbol{v}_1\cdot\boldsymbol{\nabla}\rho_1 + \rho_1\operatorname{div}\boldsymbol{v}_1 - \rho\,d_{ij}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) - \frac{\rho_1\rho_2}{\rho}d_{ij}^{(T)}\nabla_i\nabla_j T = 0 \tag{68}$$

$$\text{or}\quad \dot{\rho}_2 + \boldsymbol{v}_2\cdot\boldsymbol{\nabla}\rho_2 + \rho_2\operatorname{div}\boldsymbol{v}_2 + \rho\,d_{ij}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) + \frac{\rho_1\rho_2}{\rho}d_{ij}^{(T)}\nabla_i\nabla_j T = 0 \tag{69}$$

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the entropy dynamics (heat conduction equation)

$$\dot{\sigma} + v_i \nabla_i \sigma - \kappa_{ij} \nabla_i \nabla_j T - \frac{\rho_1 \rho_2}{\rho} d_{ij}^{(T)} \nabla_i \nabla_j (\mu_1 - \bar{\mu}_2) = 0 \qquad (70)$$

the nematic director dynamics

$$\dot{n}_i + v_j^{(2)} \nabla_j n_i - \lambda_{ijk} \nabla_j v_k^{(2)} - \frac{\rho_1}{\rho_2} \lambda_{ijk} w_k \nabla_j \phi + \gamma_1^{-1} \delta_{ij}^{\perp} \bar{h}_j = 0. \qquad (71)$$

There is a (nonlinear) coupling to the concentration variable, which is not possible in the 1-fluid description.

For the momentum balance of the two different species we get

$$\rho_1 \dot{v}_i^{(1)} + \rho_1 v_j^{(1)} \nabla_j v_i^{(1)} + \frac{\rho_1}{\rho} \nabla_i (p + \frac{1}{2} \rho_2 (\boldsymbol{v}_1^2 - \boldsymbol{v}_2^2)) + \frac{\rho_1 \rho_2}{\rho} \nabla_i (\mu_1 - \bar{\mu}_2) + \frac{\rho_1}{\rho} \nabla_j (\Psi_{kj} \nabla_i n_k)$$

$$+ \frac{\rho_1}{\rho} \bar{h}_j \nabla_i n_j + \frac{\rho_1}{\rho_2} \lambda_{kji} \bar{h}_k \nabla_j \phi + \xi_{ij} \rho_1 \rho_2 w_j - \nu_{ijkl}^{(1)} \nabla_j \nabla_l v_k^{(1)} = 0 \qquad (72)$$

$$\rho_2 \dot{v}_i^{(2)} + \rho_2 v_j^{(2)} \nabla_j v_i^{(2)} + \frac{\rho_2}{\rho} \nabla_i (p - \frac{1}{2} \rho_1 (\boldsymbol{v}_1^2 - \boldsymbol{v}_2^2)) - \frac{\rho_1 \rho_2}{\rho} \nabla_i (\mu_1 - \bar{\mu}_2) + \frac{\rho_2}{\rho} \nabla_j (\Psi_{kj} \nabla_i n_k)$$

$$- \frac{\rho_1}{\rho} \bar{h}_j \nabla_i n_j - \frac{\rho_1}{\rho_2} \lambda_{kji} \bar{h}_k \nabla_j \phi - \nabla_j (\lambda_{kji} \bar{h}_k) - \xi_{ij} \rho_1 \rho_2 w_j - \nu_{ijkl}^{(2)} \nabla_j \nabla_l v_k^{(2)} = 0 \qquad (73)$$

Note that although we made the approximation that the linear orientational-elastic stress does only act on fluid 2, there are inevitably nonlinear contributions to fluid 1, too. There is also a (nonlinear) coupling of fluid 1 to the concentration, if nematic distortions ($\bar{h}_i \neq 0$) are present. In (72,73) cross-viscosities have been neglected (cf. Appendix).

In order to facilitate actual calculations we also give eqs.(72,73) as dynamic equations for the total momentum and for the relative velocity

$$\rho \dot{v}_i + \nabla_i p + \rho \nabla_j \Big(v_i v_j + \phi(1 - \phi) w_i w_j \Big) + \nabla_j \Big(\Psi_{kj} \nabla_i n_k - \lambda_{kji} \bar{h}_k \Big)$$

$$- \nu_{ijkl} \nabla_j \nabla_l v_k = 0 \quad (74)$$

$$\dot{w}_i + \Big(v_j + (1 - 2\phi) w_j \Big) \nabla_j w_i + \nabla_i \Big(\mu_1 - \bar{\mu}_2 + \boldsymbol{v} \cdot \boldsymbol{w} + (\frac{1}{2} - \phi) \boldsymbol{w}^2 \Big) + \rho \xi_{ij} w_j$$

$$+ \frac{1}{\rho_2} \bar{h}_j \nabla_i n_j + \nabla_j \Big(\frac{1}{\rho_2} \lambda_{kji} \bar{h}_k \Big) = 0 \quad (75)$$

Note that the approximation for the viscosities made in (74,75) is not compatible with that used in (72,73); their interrelation is discussed in the Appendix.

Due to the incompressibility condition the pressure is no longer an independent variable nor is it given by the other variables (i.e. (5) or (9) cannot be used), but it serves as an auxiliary quantity to ensure the incompressibility for all times, i.e. div$\dot{\boldsymbol{v}} = 0$, which leads to the very complicated condition that determines δp

$$\Delta p = -\nabla_i \nabla_j (\rho_1 v_i^{(1)} v_j^{(1)} + \rho_2 v_i^{(2)} v_j^{(2)}) - \nabla_i \nabla_j (\Psi_{kj} \nabla_i n_k) \qquad (76)$$

$$+ \lambda \nabla_i \nabla_j (\delta_{kj}^{\perp} n_i \bar{h}_k) + \nu_{ijkl} \nabla_i \nabla_j \nabla_l v_k$$

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Although δp does not show up in the dynamical equations, it is still present in boundary conditions etc. and it contains combinations of the viscosities different from those present in the incompressible dynamical equations.[4] In contrast to 1-fluid descriptions for simple fluids, where the incompressibility condition leads to a considerable mathematical simplification, this is no longer the case for a 2-fluid description due the complicated form of (76), even if incompressibility is a very good approximation in physical terms.

Of the statics (28–33) only the following equations remain

$$\delta T = T C_V^{-1} \delta\sigma + \alpha_\phi^{-1}\delta\phi \tag{77}$$

$$\delta(\mu_1 - \bar{\mu}_2) = \rho^{-1}\kappa_\phi^{-1}\,\delta\phi + \rho^{-1}\alpha_\phi^{-1}\,\delta\sigma \tag{78}$$

with $\delta\phi = \rho^{-1}\delta\rho_1 = -\rho^{-1}\delta\rho_2$, while (39) and (40) are unchanged. Note that $\delta\mu$ is not needed, but follows from δp via eq. (9).

8   Isotropic Viscoelastic Fluids

In this section we discuss the 2-fluid description of isotropic viscoelastic fluids by choosing a Newtonian fluid as fluid 1 and an elastic medium as fluid 2. The latter can be a permanent network (showing e.g. diffusion) or a temporary one relaxing on a finite time scale. The considerations for setting up a complete nonlinear 2-fluid description for such systems is quite similar to that of the 2-fluid nematic discussed in detail in the previous sections - only that the nematic degree of freedom $\nabla_j n_i$ has to be replaced by the Eulerian strain tensor U_{ij}, which we use to describe the elastic degree of freedom. In the following we present an abbreviated discussion, starting with the general energy expression.

8.1   Thermodynamics

In analogy with the development in section 2, the general energy expression for an isotropic elastomer network immersed in a Newtonian solvent is given by

$$E = \epsilon V = \int \epsilon\, dV = E(M_1,\, M_2,\, V,\, \boldsymbol{G_1},\, \boldsymbol{G_2},\, S,\, M_2 U_{ij}) \tag{79}$$

from which the conjugate quantities are derived. The elastic stress, conjugate to the strain, is $\Phi'_{ij} = \partial E/\partial(M_2 U_{ij}) = \partial\epsilon/\partial(\rho_2 U_{ij}) \equiv \rho_2^{-1}\Phi_{ij}$, while the definitions of the other conjugates (2) remain unchanged (except that they are to be taken at constant U_{ij} rather than constant $\nabla_j n_i$). With the new definition $\bar{\mu}_2 = \mu_2 + \rho^{-1}\Phi_{ij}U_{ij}$ the expressions for the

---

[4]Incompressibility 'reduces' the number of independent components of the viscosity tensor from five to three (in the uniaxial case), only if a redefinition of the pressure is done, cf. [28] p.41f and [33]

pressure (3,7) and the relations of the different sets of conjugates (10) remain unchanged, while the Gibbs and Gibbs-Duhem relations read

$$
\begin{aligned}
d\epsilon &= Td\sigma + \mu_1\,d\rho_1 + \bar{\mu}_2\,d\rho_2 + \boldsymbol{v}_1\cdot d\boldsymbol{g}_1 + \boldsymbol{v}_2\cdot d\boldsymbol{g}_2 + \Phi_{ij}\,dU_{ij} & (80)\\
&= T\,d\sigma + \Pi\,d\phi + \mu\,d\rho + \boldsymbol{v}\cdot d\boldsymbol{g} + \boldsymbol{m}\cdot d\boldsymbol{w} + \Phi_{ij}\,dU_{ij} & (81)\\
dp &= \sigma\,dT + \rho_1\,d\mu_1 + \rho_2\,d\bar{\mu}_2 + \boldsymbol{g}_1\cdot d\boldsymbol{v}_1 + \boldsymbol{g}_2\cdot d\boldsymbol{v}_2 - \Phi_{ij}\,dU_{ij} & (82)\\
&= \sigma\,dT + \rho\,d\mu + \boldsymbol{g}\cdot d\boldsymbol{v} - \boldsymbol{m}\cdot d\boldsymbol{w} - \Phi_{ij}\,dU_{ij} & (83)
\end{aligned}
$$

Rotational invariance of the Gibbs relation (80,81) leads to the condition

$$
U_{ik}\Phi_{kj} = U_{jk}\Phi_{ki} \tag{84}
$$

which, as is seen later on, ensures the stress tensor to be symmetric.

8.2 Statics

The conjugate quantities defined by the Gibbs relation (80,81) are linked to the variables by a set of phenomenological equations containing static susceptibilities as parameters. This constitutes the static part of the hydrodynamics. Instead of the nematic molecular fields h_i and ψ_{ij} we now have the elastic stress Φ_{ij} as conjugate field. As a symmetric 2-rank tensor it consists of a scalar quantity, the trace Φ_{ii} and the deviator $\Phi_{ij}^{(0)} = \Phi_{ij} - (1/3)\delta_{ij}\Phi_{kk}$. Being a scalar Φ_{kk} can couple to the other scalar variables like densities, concentration or entropy, just like the 3 other scalar conjugates $\{T, \Pi, \mu \text{ or } \mu_1, \mu_2\}$ by

$$
\begin{aligned}
\delta T &= \frac{T}{C_V}\delta\sigma + \frac{1}{\rho\alpha_1}\delta\rho_1 + \frac{1}{\rho\alpha_2}\delta\rho_2 + \frac{1}{\alpha_3}U_{kk} & (85)\\
&= \frac{T}{C_V}\delta\sigma + \frac{1}{\alpha_\phi}\delta\phi + \frac{1}{\rho\alpha_\rho}\delta\rho + \frac{1}{\alpha_3}U_{kk}\\
\mu_1 &= \frac{1}{\rho^2\kappa_1}\delta\rho_1 + \frac{1}{\rho^2\kappa_3}\delta\rho_2 + \frac{1}{\rho\alpha_1}\delta\sigma + \frac{1}{\rho\kappa_4}U_{kk} & (86)\\
\Pi &= \frac{1}{\kappa_\phi}\delta\phi + \frac{1}{\rho\kappa_\pi}\delta\rho + \frac{1}{\alpha_\phi}\delta\sigma + \frac{1}{\kappa_u}U_{kk} + \boldsymbol{w}\cdot\boldsymbol{g} + \rho\boldsymbol{w}^2(1-2\phi) & (87)\\
\bar{\mu}_2 &= \frac{1}{\rho^2\kappa_2}\delta\rho_2 + \frac{1}{\rho^2\kappa_3}\delta\rho_1 + \frac{1}{\rho\alpha_2}\delta\sigma + \frac{1}{\rho\kappa_5}U_{kk} & (88)\\
\mu &= \frac{1}{\rho^2\kappa_\mu}\delta\rho + \frac{1}{\rho\kappa_\pi}\delta\phi + \frac{1}{\rho\alpha_\rho}\delta\sigma + \frac{1}{\rho\kappa_\rho}U_{kk} + \boldsymbol{w}^2\phi(1-\phi) & (89)\\
\Phi_{kk} &= c_l U_{kk} + \frac{1}{\alpha_3}\delta\sigma + \frac{1}{\rho\kappa_4}\delta\rho_1 + \frac{1}{\rho\kappa_5}\delta\rho_2 & (90)\\
&= c_l U_{kk} + \frac{1}{\alpha_3}\delta\sigma + \frac{1}{\rho\kappa_u}\delta\phi + \frac{1}{\rho\kappa_\rho}\delta\rho\\
\Phi_{ij}^{(0)} &= c_{tr}(U_{ij} - \frac{1}{3}\delta_{ij}U_{kk}) & (91)
\end{aligned}
$$

where – in addition to (34–38)

$$\kappa_\rho^{-1} = \phi\kappa_4^{-1} + (1-\phi)\kappa_5^{-1} \tag{92}$$

$$\kappa_u^{-1} = \kappa_4^{-1} - \kappa_5^{-1} \tag{93}$$

involving 2 new generalized compressibilities $\kappa_{4,5}$ or $\kappa_{u,\rho}$ and one expansion coefficient α_3 related to the trace of the elastic strain U_{kk}. It should be noted that for real solids at finite temperatures $U_{kk} \neq \delta\rho/\rho$ in contrast to ideal elasticity theory. The reason are the point defects, which allow not only the dissipative motion described above, but also static temperature and pressure changes due to U_{kk} even at constant density. The new static susceptibilities c_l and c_{tr} are the usual elastic moduli of Hooke's law. The longitudinal one is related (in addition to the compressibility κ_μ) to the sound velocity. The transverse modulus leads to transverse sound, which is however relaxing due to (104) if ζ_l and ζ_{tr} are not zero.

8.3 Dynamics

The dynamical equations for the elastomeric and solvent degrees of freedom are

$$\dot\rho + \nabla_j \rho\, v_j = 0 \tag{94}$$

$$\dot\phi + v_j\nabla_j\phi + \rho^{-1}\nabla_i\left(\rho\phi(1-\phi)w_i + j_i^{(1)}\right) = 0 \tag{95}$$

$$\dot\epsilon + \nabla_j(\epsilon + p)v_j + \nabla_i j_i^{(\epsilon)} = 0 \tag{96}$$

$$\dot\sigma + \nabla_j(\sigma v_j + j_i^{(\sigma,rev)} + j_i^{(\sigma,dis)}) = R/T \tag{97}$$

$$\dot w_i + v_j\nabla_j w_i + \nabla_i\Pi + X_i^{(rev)} + X_i^{(dis)} = 0 \tag{98}$$

$$\dot g_i + \nabla_j g_i v_j + \nabla_i p + \nabla_j(-\Phi_{ij} + \Phi_{jk}U_{ik} + \Phi_{ik}U_{jk} + \sigma_{ij}^{(rev)} + \sigma_{ij}^{(dis)}) = 0 \tag{99}$$

$$\dot U_{ij} + v_k\nabla_k U_{ij} + U_{kj}\nabla_i v_k + U_{ki}\nabla_j v_k - A_{ij} + Z_{ij}^{(rev)} + Z_{ij}^{(dis)} = 0 \tag{100}$$

the first 5 equations have the same form as before (but different phenomenological currents, see below). In the dynamic equation for the strain (100) there are nonlinear couplings to the velocity gradient that have the form of the so-called lower convected derivative [34, 35] and the appropriate counter terms show up in the stress tensor as additions to the phenomenological parts. In all dynamic equations the convective velocity chosen is the mean velocity $\boldsymbol{v}$, since this allows a simple and thermodynamically consistent way of writing the equations. However, as in the case discussed previously there are phenomenological terms in the reversible currents that allow a different choice of the convective velocities (see below).

8.4 Currents

Following the previous development, we can establish the reversible and dissipative currents, and their constraints.

For the phenomenological parts of the currents there is the condition

$$R = -j_i^{(\sigma,*)}\nabla_i T + \Pi \; \nabla_i j_i^{(1)} - \sigma_{ij}^{(*)}\nabla_j v_i + \Phi_{ij} Z_{ij}^{(*)} + m_i \, X_i^{(*)} \geq 0 \tag{101}$$

with the equal sign ($>$ sign) for $* = rev$ ($* = dis$), respectively.

The dissipative parts of the currents introduced above can again be deduced from a dissipation function that reads in bilinear approximation

$$\begin{aligned}
2R \;=\; & \kappa(\boldsymbol{\nabla}T)^2 + D(\boldsymbol{\nabla}\Pi)^2 + 2D^{(T)}(\boldsymbol{\nabla}T)\cdot(\boldsymbol{\nabla}\Pi) + \xi' \boldsymbol{m}^2 \\
& +\zeta_{ijkl}\Phi_{ij}\Phi_{kl} + \xi_{ijklmn}(\nabla_i\Phi_{jk})(\nabla_l\Phi_{mn}) + \nu_{ijkl}(\nabla_j v_i)(\nabla_l v_k) \\
& +\nu_{ijkl}^{(c)}\left((\nabla_j v_i)(\nabla_l m_k) + (\nabla_j m_i)(\nabla_l v_k)\right) + \nu_{ijkl}^{(w)}(\nabla_j m_i)(\nabla_l m_k)
\end{aligned} \tag{102}$$

where all 4-rank material tensors have the form $\nu_{ijkl} = \nu_l\delta_{ij}\delta_{kl} + (1/2)\nu_{tr}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - (2/3)\delta_{ij}\delta_{kl}))$ and ξ_{ijklmn} contains 4 parameters ξ_{1-4}. In the ζ-tensor ζ_l and ζ_{tr} are describing the relaxation of elastic strains and the ξ_{1-4} give rise to vacancy diffusion as can be seen in the following expressions

$$\begin{aligned}
j_i^{(\sigma,dis)} \;&=\; -(\partial R)/(\partial\nabla_i T) = -\kappa\nabla_i T - \rho\,\phi(1-\phi)\,d^{(T)}\nabla_i\Pi \tag{103} \\
Z_{ij}^{(dis)} \;&=\; (\partial R)/(\partial\Phi_{ij}) = \zeta_{ijkl}\Phi_{kl} - \nabla_k(\xi_{kijlmn}\nabla_l\Phi_{mn}) \tag{104} \\
\sigma_{ij}^{(dis)} \;&=\; -(\partial R)/(\partial\nabla_j v_i) = -\nu_{ijkl}\nabla_l v_k - \nu_{ijkl}^{(c)}\nabla_l m_k \tag{105} \\
X_i^{(dis)} \;&=\; (\partial R)/(\partial m_i) = \xi'\,m_i - \nabla_j\left(\nu_{ijkl}^{(w)}\nabla_l w_k + \nu_{ijkl}^{(c)}\nabla_l v_k\right) \tag{106} \\
j_i^{(1,dis)} \;&=\; -(\partial R)/(\partial\nabla_i\Pi) = -\rho\,d\nabla_i\Pi - \rho\,\phi(1-\phi)\,d^{(T)}\nabla_i T \tag{107}
\end{aligned}$$

where diffusion and thermodiffusion is written in the usual way with $D = \rho d$ and $D^{(T)} = \rho\phi(1-\phi)d^{(T)}$. For a permanent network that does not relax, the relaxation parameters vanish ($\zeta_l = \zeta_{tr} = 0$). For the reversible parts of the currents we find

$$\begin{aligned}
Z_{ij}^{(rev)} \;&=\; \lambda^{(U)}(\nabla_i m_j + \nabla_j m_i) + \beta_7(U_{kj}\nabla_i m_k + U_{ki}\nabla_j m_k) + \beta_6 m_k\nabla_k U_{ij} \tag{108} \\
\sigma_{ij}^{(rev)} \;&=\; 2\beta_2\,m_i\,w_j \tag{109} \\
X_i^{(rev)} \;&=\; 2\nabla_j(\lambda^{(U)}\Phi_{ij}) + \beta\,\nabla_i T + \gamma\nabla_i\Pi - \beta_6\Phi_{kj}\nabla_i U_{kj} + \beta_2 w_j(\nabla_j v_i + \nabla_i v_j) \\
& \quad +\nabla_j\beta_7(\Phi_{kj}U_{ik} + \Phi_{ki}U_{jk}) + \beta_3 m_j(\nabla_j w_i - \nabla_i w_j) + \beta_4 w_j(\nabla_j v_i - \nabla_i v_j) \tag{110} \\
j_i^{(\sigma,rev)} \;&=\; \beta\,m_i \tag{111} \\
j_i^{(1,rev)} \;&=\; \gamma\,m_i \tag{112}
\end{aligned}$$

8.5 Convection, Stress, and Concentration Dynamics

As in the case of the 2-fluid nematics the velocities which with the variables are convected can be tuned by choosing special values for the coefficients β_n and γ. E.g. for $\gamma = 0$

the densities $\rho_{1,2}$ are convected with $\boldsymbol{v}_{1,2}$, respectively (and the total density ρ and the concentration ϕ with $\boldsymbol{v}$ and $(1/\rho)(\rho_2\boldsymbol{v}_1 + \rho_1\boldsymbol{v}_2)$, respectively), while for $\gamma = -1$ all 4 quantities are convected with $\boldsymbol{v}$. Similarly, for $\beta_4 = 1/2 = \beta_2$ and $\beta_3 = (1/\rho_1) - (1/\rho_2)$ the momenta $\boldsymbol{g_{1,2}}$ are convected with $\boldsymbol{v}_{1,2}$ (and the total momentum $\boldsymbol{g}$ and the relative velocity $\boldsymbol{w}$ with $\boldsymbol{v}$ and $(1/\rho)(\rho_2\boldsymbol{v}_1 + \rho_1\boldsymbol{v}_2)$, respectively). For $\beta_6 = -1/\rho_2$ the strain U_{ij} is convected with $\boldsymbol{v}_2$ and for $\beta_7 = -1/\rho_2$ the lower convected derivative contributions in (100) effectively come with $\boldsymbol{v}_2$ (producing an additional cubic term in (110) $\sim \nabla_i\rho_2$, which can be neglected as other cubic terms). Even the convection of the entropy can be tuned by choosing $\beta \equiv \beta_0 + \beta_{00}\sigma$ where $\beta_{00} = 1/\rho_1, = 0, = -1/\rho_2$ leads to the convective velocity to be $\boldsymbol{v}_1$, $\boldsymbol{v}$, $\boldsymbol{v}_2$, respectively.

The distribution of the elastic stress among the two fluids is governed by the coefficient $\lambda^{(U)}$. For, respectively, $2\lambda^{(U)} = 1/\rho_2, = -1/\rho_1$, or $= 0$, the elastic stress is carried by fluid 2, fluid 1, or is equally distributed between them.

As in the case of 2-fluid nematics we can linearize and Fourier transform the dynamic equations, thus eliminating $\boldsymbol{w}$ from e.g. the concentration dynamics. Neglecting fourth order gradient terms we get

$$i\omega\phi - d^{eff}\Delta\Pi - \frac{\rho_1\rho_2}{\rho^2}d^{(T)\,eff}\Delta T - 2\lambda^{(\phi)}\nabla_i\nabla_j\Phi_{ij} = 0 \tag{113}$$

with frequency dependent effective diffusion and thermo-diffusion coefficients

$$d^{eff} \;=\; d + \frac{\rho_1\rho_2}{\rho^2}\frac{(\gamma+1)^2}{\rho\,\xi + i\omega} \tag{114}$$

$$d^{(T)\,eff} \;=\; d^{(T)} + \frac{\beta(\gamma+1)}{\rho\,\xi + i\omega} \tag{115}$$

and the dynamic coupling to the elastic degree of freedom by

$$\lambda^{(\phi)} = \frac{\rho_1\rho_2}{\rho}\lambda^{(U)}\frac{1+\gamma}{\rho\xi + i\omega} \tag{116}$$

Again these possible additions to the concentration dynamics, however, depend on the choices for the convection velocities as well as on the way how the nematic stress has been divided among the two fluids. Assuming the densities $\rho_{1,2}$ to be convected with the mean velocity $\boldsymbol{v}$ (implying $\gamma = -1$) the additional contributions to diffusion and thermo-diffusion are all zero as well as the coupling to the nematic director. On the other hand, for $\rho_{1,2}$ to be convected with $\boldsymbol{v}_{1,2}$ respectively (e.g. $\gamma = 0$), both, diffusion and thermo-diffusion show a dispersion step around $\omega \approx \rho\,\xi$. For the elastic stress division among fluid 1 and 2 according to the ratio ρ_1/ρ_2 (implying $\lambda_1^{(U)} = 0$) there is no dynamic influence of the elastic degree of freedom on the concentration, while for any other choice there is one. In particular, if only fluid 2 carries elastic stress ($\lambda_1^{(U)} = 1/\rho_2$), this dynamic coupling is given by $\lambda^{(\phi)} = \rho_1/\rho^2\xi$ for strong friction ($\rho\xi \gg \omega$).

8.6 Simplified elastomeric two-fluid equations

In the preceding sections we have derived the most general and complete set of 2-fluid equations. These equations are for most purposes unnecessarily complicated and can be simplified using reasonable assumptions. Starting from the correct general equations such assumptions, clearly spelled out, lead to controlled approximations and to a set of 2-fluid equations, whose limitations and implicit assumptions are clear and well defined in contrast to most ad-hoc approaches.

Here we want to display explicitly 2-fluid hydrodynamics under the following assumptions,

a) convection with natural velocities (for U_{ij}, $\boldsymbol{g}_2$, ρ_2 and $\boldsymbol{g}_1$, ρ_1 this is $\boldsymbol{v}_2$ and $\boldsymbol{v}_1$, respectively, or explicitly $\beta_7 = -\rho_2^{-1} = \beta_6$, $\beta_2 = \beta_4 = 1/2$, $\beta_3 = \rho_1^{-1} - \rho_2^{-1}$, $\gamma = 0$);

b) the linearized elastic force acts on the elastomeric fluid (index 2) only (i.e. $2\lambda^{(U)} = \rho_2^{-1}$);

c) global incompressibility, $\delta\rho = 0$ (i.e. $\delta\rho_1 = -\delta\rho_2$);

d) linearizing the phenomenological dissipative currents, but keeping quadratic nonlinearities otherwise.

Then the following set of equations is obtained:

The incompressibility condition

$$0 \;=\; \operatorname{div}\boldsymbol{v} \tag{117}$$

$$\text{or}\qquad 0 \;=\; \boldsymbol{w}\cdot\boldsymbol{\nabla}\rho_1 + \rho_1\operatorname{div}\boldsymbol{v}_1 + \rho_2\operatorname{div}\boldsymbol{v}_2 \tag{118}$$

$$\text{or}\qquad 0 \;=\; \boldsymbol{w}\cdot\boldsymbol{\nabla}\phi + \phi\operatorname{div}(1-\phi)\boldsymbol{w} - (1-\phi)\operatorname{div}\phi\boldsymbol{w} \tag{119}$$

the concentration dynamics

$$\dot{\phi} + \nabla_i\left(\phi v_i + \phi(1-\phi)w_i\right) - d_{ij}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) - \phi(1-\phi)d_{ij}^{(T)}\nabla_j\nabla_i T \;=\; 0 \tag{120}$$

$$\text{or}\qquad \dot{\rho}_1 + \boldsymbol{v}_1\cdot\boldsymbol{\nabla}\rho_1 + \rho_1\operatorname{div}\boldsymbol{v}_1 - \rho\,d_{ij}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) - \frac{\rho_1\rho_2}{\rho}d_{ij}^{(T)}\nabla_i\nabla_j T \;=\; 0 \tag{121}$$

$$\text{or}\qquad \dot{\rho}_2 + \boldsymbol{v}_2\cdot\boldsymbol{\nabla}\rho_2 + \rho_2\operatorname{div}\boldsymbol{v}_2 + \rho\,d_{ij}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) + \frac{\rho_1\rho_2}{\rho}d_{ij}^{(T)}\nabla_i\nabla_j T \;=\; 0 \tag{122}$$

the entropy dynamics (heat conduction equation)

$$\dot{\sigma} + v_i\nabla_i\sigma + \frac{\beta}{\rho}\nabla_i(\rho_1\rho_2 w_i) - \kappa_{ij}\nabla_i\nabla_j T - \frac{\rho_1\rho_2}{\rho}d_{ij}^{(T)}\nabla_i\nabla_j(\mu_1 - \bar{\mu}_2) = 0 \tag{123}$$

the elasticity dynamics

$$\dot{U}_{ij} \;+\; v_k^{(2)}\nabla_k U_{ij} - \frac{1}{2}(\nabla_j v_i^{(2)} + \nabla_i v_j^{(2)}) - \frac{\rho_1}{2}(w_i\nabla_j + w_j\nabla_i)\ln\frac{\rho_2}{\rho} + U_{ki}\nabla_j v_k^{(2)} + U_{kj}\nabla_i v_k^{(2)}$$
$$+\; \zeta_l\delta_{ij}\Phi_{kk} + \zeta_{tr}(\Phi_{ij} - \frac{1}{3}\delta_{ij}\Phi_{kk}) - \xi_1\delta_{ij}\Delta\Phi_{kk} - \xi_2\Delta\Phi_{ij} - \xi_3(\nabla_i\nabla_j\Phi_{kk} + \delta_{ij}\nabla_k\nabla_l\Phi_{kl})$$
$$-\; \xi_4(\nabla_i\nabla_k\Phi_{jk} + \nabla_j\nabla_k\Phi_{ik}) = 0 \tag{124}$$

There are nonlinear couplings to the concentration variable (the cubic one has been suppressed), which are not possible in a 1-fluid description.

For the momentum balance of the two different species we get

$$\rho_1\dot{v}_i^{(1)} + \rho_1 v_j^{(1)}\nabla_j v_i^{(1)} + \frac{\rho_1}{\rho}\nabla_i(p + \frac{1}{2}\rho_2(\boldsymbol{v}_1^2 - \boldsymbol{v}_2^2)) + \frac{\rho_1\rho_2}{\rho}\nabla_i(\mu_1 - \bar{\mu}_2) + \frac{\rho_1}{\rho}\Phi_{kj}\nabla_i U_{kj}$$
$$- \rho_1\Phi_{ij}\nabla_j \ln\frac{\rho_2}{\rho} + \frac{\rho_1\rho_2}{\rho}\beta\nabla_i T + \xi_{ij}\rho_1\rho_2 w_j - \nu_{ijkl}^{(1)}\nabla_j\nabla_l v_k^{(1)} - \nu_{ijkl}^{(12)}\nabla_j\nabla_l v_k^{(2)} = 0 \quad (125)$$

$$\rho_2\dot{v}_i^{(2)} + \rho_2 v_j^{(2)}\nabla_j v_i^{(2)} + \frac{\rho_2}{\rho}\nabla_i(p - \frac{1}{2}\rho_1(\boldsymbol{v}_1^2 - \boldsymbol{v}_2^2)) - \frac{\rho_1\rho_2}{\rho}\nabla_i(\mu_1 - \bar{\mu}_2) - \frac{\rho_1}{\rho}\Phi_{kj}\nabla_i U_{kj}$$
$$+ \rho_1\Phi_{ij}\nabla_j \ln\frac{\rho_2}{\rho} - \frac{\rho_1\rho_2}{\rho}\beta\nabla_i T - \nabla_j\Phi_{ij} + \nabla_j(\Phi_{jk}U_{ik} + \Phi_{ik}U_{jk}) - \xi_{ij}\rho_1\rho_2 w_j$$
$$- \nu_{ijkl}^{(2)}\nabla_j\nabla_l v_k^{(2)} - \nu_{ijkl}^{(12)}\nabla_j\nabla_l v_k^{(1)} = 0 \quad (126)$$

Note that although we made the approximation that the linear elastic stress does only act on fluid 2, there are inevitably nonlinear contributions to fluid 1, too. There is also a (nonlinear) coupling of fluid 1 to the concentration, if elastic distortions are present.

The different approximations for the viscosities are discussed in the Appendix.

In order to facilitate actual calculations we also give eqs.(125,126) as dynamic equations for the total momentum and for the relative velocity

$$\rho\dot{v}_i + \nabla_i p + \nabla_j\left(\rho v_i v_j + \frac{\rho_1\rho_2}{\rho}w_i w_j\right) - \nabla_j\Phi_{ij}$$
$$+ 2\nabla_j(\Phi_{jk}U_{ik}) - \nu_{ijkl}\nabla_j\nabla_l v_k - \frac{\rho_1\rho_2}{\rho}\nu_{ijkl}^{(c)}\nabla_j\nabla_l w_k) = 0 \quad (127)$$

$$\dot{w}_i + \left(v_j + \frac{\rho_2 - \rho_1}{\rho}w_j\right)\nabla_j w_i + \nabla_i\left(\mu_1 - \bar{\mu}_2 + \boldsymbol{v}\cdot\boldsymbol{w} + \frac{\rho_2 - \rho_1}{2\rho}\boldsymbol{w}^2\right) + \rho\xi_{ij}w_j + \nabla_j\frac{1}{\rho_2}\Phi_{ij}$$
$$+ \frac{1}{\rho_2}\Phi_{kj}\nabla_i U_{kj} - \frac{2}{\rho_2}\nabla_j(\Phi_{kj}U_{ik}) - \frac{\rho_1\rho_2}{\rho}\nu_{ijkl}^{(m)}\nabla_l\nabla_j w_k - \nu_{ijkl}^{(c)}\nabla_j\nabla_l v_k = 0 \quad (128)$$

In order to conserve the global incompressibility condition for all times, i.e. $\text{div}\dot{\boldsymbol{v}} = 0$, the pressure has to fulfill the relation

$$\Delta p = -\nabla_i\nabla_j(\rho_1 v_i^{(1)}v_j^{(1)} + \rho_2 v_i^{(2)}v_j^{(2)}) + \nabla_i\nabla_j\Phi_{ij} - \nabla_i\nabla_j(\Phi_{kj}U_{ik} + \Phi_{ik}U_{jk})$$
$$+ \nu_{ijkl}\nabla_i\nabla_j\nabla_l v_k + \rho_1\rho_2\rho^{-1}\nu_{ijkl}^{(c)}\nabla_i\nabla_j\nabla_l w_k \quad (129)$$

In contrast to 1-fluid descriptions for simple fluids, where the incompressibility condition leads to a considerable mathematical simplification, this is no longer the case for a 2-fluid description due the complicated form of (129), even if incompressibility is a very good approximation in physical terms. In particular, Δp is not only connected to compressions (U_{kk}), but also to shear deformations, even in linear order.

Of the statics (85–90) only the following equations remain

$$\delta T = TC_V^{-1}\delta\sigma + \alpha_\phi^{-1}\delta\phi + \alpha_3^{-1}U_{kk} \quad (130)$$
$$\delta(\mu_1 - \bar{\mu}_2) = \rho^{-1}\kappa_\phi^{-1}\delta\phi + \rho^{-1}\alpha_\phi^{-1}\delta\sigma + \kappa_u^{-1}U_{kk} \quad (131)$$
$$\Phi_{kk} = c_l U_{kk} + \alpha_3^{-1}\delta\sigma + \rho^{-1}\kappa_u^{-1}\delta\phi \quad (132)$$

with $\delta\phi = \rho^{-1}\delta\rho_1 = -\rho^{-1}\delta\rho_2$, while eq.(91) remains unchanged. Note that $\delta\mu$ is not needed, but follows from δp via eq. (83).

9   Discussion

Within the general framework of hydrodynamics and thermodynamics we have set up a consistent nonlinear 2-fluid description of complex fluids, in particular for lyotropic nematic liquid crystals and polymer solutions or swollen elastomers. Such a general theory determines the frame for any ad-hoc model, which has to be a special case of the general one. The comparison with the general theory also reveals implicit and explicit assumptions, approximations and possible generalizations of a given model. A simple or "natural" choice in a given model may not be mandatory, but rather imply a presumption.

Quite generally we find that neither the velocity, with which a certain variable is convected, nor the stress division between the different fluids can be determined by general principles, but is rather system or material dependent. On the other hand, there are certain restrictions and interrelations among the convective velocities and other physical effects that limit the possible choices. For the two densities ρ_1, ρ_2 e.g., the natural choice for the convection velocities seems to be their native velocities $\boldsymbol{v}_1$ and $\boldsymbol{v}_2$, respectively. This implies that the total density is convected with the mean velocity $\boldsymbol{v}$ (as required by mass transport), while the concentration ϕ is convected with $(1/\rho)(\rho_2\boldsymbol{v}_1 + \rho_1\boldsymbol{v}_2)$. Another obvious choice would be the mean velocity as convection velocity for both, the total density as well as the concentration implying that also ρ_1 and ρ_2 are convected with $\boldsymbol{v}$. However, the actual convection velocity depends on the value of the material dependent (reactive) flow parameters $\gamma_\perp$ and $\gamma_\parallel$, defined in eq. (51).

For the nematic degree of freedom the convective velocity again depends on a material parameter (β_1 defined in eq.(47)) and is not necessarily equal to $\boldsymbol{v}_2$ (if fluid 2 is the nematogen). However, the value of β_1 influences also the flow alignment of the director (and the back flow due to director reorientation), which can be measured in shear flow experiments. In the case of visco-elastic and elastic media, which are described by a dynamic equation for the (Eulerian) strain tensor U_{ij}, there are two velocities involved. One is the usual convection velocity ($v_k\nabla_k U_{ij}$) and the other one occurs in the "lower convected" part ($U_{kj}\nabla_i v_k + U_{ki}\nabla_j v_k$). There is no fundamental reason for the two to be equal and their actual value depends on the (reactive) flow parameters β_6 and β_7, respectively, defined in eqs.(108, 110).

For the evolution equations of the momenta special care has to be taken to get a description, which is compatible with general laws (cf. Chapter 6). The currents and quasi-currents that enter the description in terms of either the total momentum and the velocity difference or the two individual momenta are not the same as seen in eqs.(55, 56). In the nematic case the stress division problem depends on the flow alignment parameters

as well as on the convection velocity of the director, while in the visco-elastic case the crucial material parameter $\lambda^{(U)}$, eq.(108,110), is not related to a convective velocity. The delicate question of viscosities, and approximations related to them, is discussed in detail in the appendix.

A prominent feature of the 2-fluid description is the coupling of the concentration dynamics to the velocity difference. This leads to a frequency dependent effective diffusion and thermo-diffusion, as well as a frequency dependent coupling to the nematic or the visco-elastic degree of freedom. For low frequencies these contributions to the concentration dynamics constitute additional dissipation channels, while for the short-time dynamics (below the relaxation time of the velocity difference) they are reactive.

Recently, 2-fluid descriptions of diffusion in polymeric systems have been given [36, 37] based on the GENERIC approach making use of Poisson brackets. A detailed comparison with these formulations is beyond the scope of this manuscript and will be discussed elsewhere.

Acknowledgments

Appendix

In this Appendix we discuss viscosity and viscosity-like phenomena in the 2-fluid hydrodynamics. We show that in order to get the 1-fluid limit (the binary liquid) correctly, some care has to be taken when the usual approximations are made.

If there is only one velocity present, the viscous contribution to the dissipation function is $\nu_{ijkl}(\nabla_j v_i)(\nabla_l v_k)$ with $\nu_{ijkl} = \nu_{jikl} = \nu_{ijlk} = \nu_{klij}$, which ensures that only symmetric velocity gradients contribute to dissipation. Antisymmetric velocity gradients, $\mathrm{curl}\,\boldsymbol{v}$, describe rotations. A solid body rotation ($\mathrm{curl}\,\boldsymbol{v} = const.$), however must not increase the entropy and $(\mathrm{curl}\,\boldsymbol{v})^2$ contributions are not allowed in the dissipation function. With these symmetries the viscosity tensor has 2 coefficients for the isotropic

$$\nu_{ijkl} = \nu(\delta_{jl}\delta_{ik} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}) + \zeta\delta_{ij}\delta_{kl} \tag{A.1}$$

and 5 for the nematic case

$$\nu_{ijkl} = \nu_2\left(\delta_{jl}\delta_{ik} + \delta_{il}\delta_{jk}\right) + 2(\nu_1 + \nu_2 - 2\nu_3)\,n_i n_j n_k n_l + (\nu_5 - \nu_4 + \nu_2)(\delta_{ij}n_k n_l + \delta_{kl}n_i n_j)$$
$$+ (\nu_4 - \nu_2)\,\delta_{ij}\delta_{kl} + (\nu_3 - \nu_2)(n_j n_l \delta_{ik} + n_j n_k \delta_{il} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk}) \tag{A.2}$$

In a 2-fluid description the same restrictions hold with respect to the mean velocity $\boldsymbol{v}$, since it is the conjugate to the momentum density and $\operatorname{curl}\boldsymbol{v} = const.$ still describes solid body rotations. There are no such restrictions to the relative velocity $\boldsymbol{w}$, and $\operatorname{curl}\boldsymbol{m}$ can contribute to the dissipation. The most general form for viscous dissipation in a 2-fluid description thus reads

$$
\begin{aligned}
2R^{(vis)} &= \nu_{ijkl}(\nabla_j v_i)(\nabla_l v_k) + 2\nu_{ijkl}^{(c)}(\nabla_j v_i)(\nabla_l m_k) + \nu_{ijkl}^{(w)}(\nabla_j m_i)(\nabla_l m_k) \\
&+ \nu_{ij}^{(r)}(\operatorname{curl}\boldsymbol{m})_i(\operatorname{curl}\boldsymbol{m})_j + 2\nu_{ijk}^{(d)}(\operatorname{curl}\boldsymbol{m})_i\nabla_j v_k + 2\nu_{ijk}^{(e)}(\operatorname{curl}\boldsymbol{m})_i\nabla_j m_k \quad (A.3)
\end{aligned}
$$

Note that only ν_{ijkl} has the dimension of a viscosity, while $\nu_{ijkl}^{(c)}$ and $\nu_{ijk}^{(d)}$ are kinematic viscosities, while $\nu_{ijkl}^{(w)}$, $\nu_{ijk}^{(e)}$, and $\nu_{ij}^{(r)}$ are viscosities divided by ρ^2. The tensors ν and $\nu^{(w)}$ have the familiar form (A.1) or (A.2). For $\nu^{(c)}$ there is no a-priori reason for a $\nu_{ijkl}^{(c)} = \nu_{klij}^{(c)}$ symmetry, since $\boldsymbol{v}$ and $\boldsymbol{m}$ are not equivalent. However, as will be seen below, a consistent 2-fluid description is only possible, if this symmetry holds and $\nu_{ijkl}^{(c)}$ has the form (A.1) or (A.2).[5] The tensor $\nu_{ij}^{(r)} = \nu^{(r)}\delta_{ij}$ or $\nu_{ij}^{(r)} = \nu_1^{(r)}\delta_{ij} + \nu_2^{(r)}n_i n_j$ contains 1 or 2 coefficients in the isotropic and nematic case, respectively. The 3rd rank material tensors, symmetric in the last two indices $\nu_{ijk}^{(d,e)} = \nu_{ikj}^{(d,e)}$ are zero in the isotropic case and both carry one coefficient in the nematic case $\nu_{ijk}^{(d,e)} = \nu^{(d,e)}(\epsilon_{ikl}n_j n_l + \epsilon_{jkl}n_i n_l)$. For the dissipative currents this leads to

$$
\begin{aligned}
\sigma_{ij}^{(dis)} &= -\nu_{ijkl}\nabla_l v_k - \nu_{ijkl}^{(c)}\nabla_l m_k - \nu_{kji}^{(d)}(\operatorname{curl}\boldsymbol{m})_k \qquad\qquad (A.4) \\
X_i^{(dis)} &= \xi_{ij}' m_j - \nabla_j\Big(\nu_{ijkl}^{(w)}\nabla_l m_k + \nu_{klij}^{(c)}\nabla_l v_k + \nu_{kl}^{(r)}\epsilon_{kji}(\operatorname{curl}\boldsymbol{m})_l + \nu_{lpk}^{(d)}\epsilon_{lji}\nabla_p v_k \\
&\qquad\qquad + \nu_{lpk}^{(e)}\epsilon_{lji}\nabla_p m_k + \nu_{kji}^{(e)}(\operatorname{curl}\boldsymbol{m})_k\Big) \qquad\qquad (A.5)
\end{aligned}
$$

Since there is already friction $\sim m_i$, very often the viscosity-like dissipation $\sim \nabla_j m_i$ is neglected altogether ($\nu^{(c)} = \nu^{(d)} = \nu^{(e)} = \nu^{(w)} = \nu^{(r)} = 0$). Such an approximation leads to (43,44). In the strong coupling limit, where $\boldsymbol{w}$ (and $\boldsymbol{m}$) vanish, this approximation seems to be appropriate and it correctly gives the 1-fluid limit of binary mixtures. On the other hand, for two fluids only gently coupled there is no a-priori reason, why e.g. the tensor $\nu^{(c)}$ (or $\nu^{(w)}$) should be neglected compared to ν, since both terms contain gradients of $\boldsymbol{v}_1$ as well as of $\boldsymbol{v}_2$. Indeed, the dissipation function in terms of $\boldsymbol{v}_{1,2}$ reads

$$
\begin{aligned}
2R^{(vis)} &= \nu_{ijkl}^{(1)}(\nabla_j v_i^{(1)})(\nabla_l v_k^{(1)}) + 2\nu_{ijkl}^{(12)}(\nabla_j v_i^{(1)})(\nabla_l v_k^{(2)}) + \nu_{ijkl}^{(2)}(\nabla_j v_i^{(2)})(\nabla_l v_k^{(2)}) \\
&+ \frac{\rho_1^2\rho_2^2}{\rho^2}\nu_{ij}^{(r)}(\operatorname{curl}[\boldsymbol{v}_1 - \boldsymbol{v}_2])_i(\operatorname{curl}[\boldsymbol{v}_1 - \boldsymbol{v}_2])_j \\
&+ 2\frac{\rho_1\rho_2}{\rho}(\operatorname{curl}[\boldsymbol{v}_1 - \boldsymbol{v}_2])_i(\nu_{ijk}^{(d1)}\nabla_j v_k^{(1)} + \nu_{ijk}^{(d2)}\nabla_j v_k^{(2)}) \qquad (A.6)
\end{aligned}
$$

[5]Without this symmetry the form (A.1) still applies for the isotropic case, while in the nematic case an additional coefficient is present, i.e. the term $\nu_5(n_i n_j\delta_{kl} + n_k n_l\delta_{ij})$ in (A.2) splits into two different parts, $\nu_{5a}n_i n_j\delta_{kl} + \nu_{5b}n_k n_l\delta_{ij}$.

where curl $\boldsymbol{v}$ is absent in the dissipation function. Comparing (A.3) and (A.6) in harmonic approximation, i.e. neglecting cubic and quartic terms involving e.g. $(\mathrm{curl}\,\boldsymbol{v}_1)_i v_k^{(1)} \nabla_j \rho_1$, $(\boldsymbol{v}_1 \times \boldsymbol{\nabla}\rho_1)_i v_k^{(1)} \nabla_j \rho_1$, or $v_i^{(1)}(\nabla_j \rho_1)(\nabla_l v_k^{(1)})$, $v_i^{(1)} v_k^{(1)}(\nabla_j \rho_1)(\nabla_l \rho_1)$, we get

$$\nu^{(d1)} = \rho_1 \nu^{(d)} + \rho_1 \rho_2 \nu^{(e)} \tag{A.7}$$

$$\nu^{(d2)} = \rho_2 \nu^{(d)} - \rho_1 \rho_2 \nu^{(e)} \tag{A.8}$$

$$\rho^2 \nu_{ijkl}^{(1)} = \rho_1^2 \nu_{ijkl} + 2\rho_1^2 \rho_2 \nu_{ijkl}^{(c)} + \rho_1^2 \rho_2^2 \nu_{ijkl}^{(w)} \tag{A.9}$$

$$\rho^2 \nu_{ijkl}^{(2)} = \rho_2^2 \nu_{ijkl} - 2\rho_2^2 \rho_1 \nu_{ijkl}^{(c)} + \rho_1^2 \rho_2^2 \nu_{ijkl}^{(w)} \tag{A.10}$$

$$\rho^2 \nu_{ijkl}^{(12)} = \rho_1 \rho_2 \nu_{ijkl} + \rho_1 \rho_2 (\rho_2 \nu_{klij}^{(c)} - \rho_1 \nu_{ijkl}^{(c)}) - \rho_1^2 \rho_2^2 \nu_{ijkl}^{(w)} \tag{A.11}$$

Again there is no a-priori reason for $\nu_{ijkl}^{(12)} = \nu_{klij}^{(12)}$. However, since the tensors ν, $\nu^{(w)}$, $\nu^{(1)}$, and $\nu^{(2)}$ do have this symmetry, eqs.(A.9,A.10) force $\nu^{(c)}$ to have it, and finally (A.11) requires also $\nu^{(12)}$ to have this symmetry and thus to be of the form (A.1,A.2).

Neglecting the curl-terms means the same in both descriptions (A.3) and (A.6), i.e. $\nu^{(r)} = \nu^{(d)} = \nu^{(d1)} = \nu^{(d2)} = 0$. For the symmetric velocity gradient terms the approximation $\nu^{(c)} = \nu^{(w)} = 0$ used in (44,45) (i.e. no $\nabla_i m_j$-terms in (A.3)) leads to $\rho_2^2 \nu^{(1)} = \rho_1^2 \nu^{(2)} = \frac{1}{2}\rho_1 \rho_2 \nu^{(12)}$ leaving only one viscous tensor independent. Neglecting only the cross-viscosity $\nu^{(c)}$ in (A.3) does not imply the cross-viscosity in (A.6) to vanish, since $\nu^{(c)} = 0$ gives $\rho\nu^{(12)} = \rho_2 \nu^{(1)} + \rho_1 \nu^{(2)}$. The opposite case $\nu^{(12)} = 0$ used in (72,73) leads to a non-zero $\rho_1 \rho_2 \nu^{(c)} = \rho_2 \nu^{(1)} - \rho_1 \nu^{(2)}$ (and $\nu = \nu^{(1)} + \nu^{(2)}$, $\nu^{(w)} = \rho_1^{-2}\nu^{(1)} + \rho_2^{-2}\nu^{(2)}$). Thus, the approximations leading to (44,45) are not compatible to those used in (72,73).

In the general case (A.6) leads to the following viscous contributions to the left hand sides of (72) and (73), respectively

$$- \nu_{ijkl}^{(1)} \nabla_j \nabla_l v_k^{(1)} - \nu_{ijkl}^{(12)} \nabla_j \nabla_l v_k^{(2)} - \frac{\rho_1 \rho_2}{\rho} \nu_{kij}^{(d1)} \nabla_j (\mathrm{curl}\,[\boldsymbol{v}_1 - \boldsymbol{v}_2])_k$$

$$-\frac{\rho_1^2 \rho_2^2}{\rho^2} \nu_{kl}^{(r)} \epsilon_{kji} \nabla_j (\mathrm{curl}\,[\boldsymbol{v}_1 - \boldsymbol{v}_2])_l - \frac{\rho_1 \rho_2}{\rho} \epsilon_{lji} \nabla_j \left(\nu_{lpk}^{(d1)} \nabla_p v_k^{(1)} + \nu_{lpk}^{(d2)} \nabla_p v_k^{(2)} \right) \tag{A.12}$$

and

$$- \nu_{ijkl}^{(2)} \nabla_j \nabla_l v_k^{(2)} - \nu_{klij}^{(12)} \nabla_j \nabla_l v_k^{(1)} - \frac{\rho_1 \rho_2}{\rho} \nu_{kij}^{(d2)} \nabla_j (\mathrm{curl}\,[\boldsymbol{v}_1 - \boldsymbol{v}_2])_k$$

$$+\frac{\rho_1^2 \rho_2^2}{\rho^2} \nu_{kl}^{(r)} \epsilon_{kji} \nabla_j (\mathrm{curl}\,[\boldsymbol{v}_1 - \boldsymbol{v}_2])_l + \frac{\rho_1 \rho_2}{\rho} \epsilon_{lji} \nabla_j \left(\nu_{lpk}^{(d1)} \nabla_p v_k^{(1)} + \nu_{lpk}^{(d2)} \nabla_p v_k^{(2)} \right) \tag{A.13}$$

In the 1-fluid limit $\nu^{(d1,d2)}$ have to vanish and $\rho(\nu^{(1)}+\nu^{(12)}) \to \rho_1 \nu$ and $\rho(\nu^{(2)}+\nu^{(12)}) \to \rho_2 \nu$, which is obtained for vanishing $\nu^{(w)}$ and $\nu^{(c)}$.

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